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Eidgenössischen Technischen Hochschule
Zürich
Single crystal of Na$_{1.7}$Os$_2$O$_{6.5}$ α-pyrochlore. The crystallization processes were carried out in a cubic anvil press at 30 kbar with different temperature regimes, starting from mixtures of alkaline metal oxides (peroxides) and OsO$_2$. As a result single crystals of the novel sodium osmate have been obtained.

This annual report was edited by: S. Gustavsson
The research results of the laboratory for solid state physics at ETH Zurich are presented in the following report. Numerous research results have been published and presented at international conferences.

Prof. H. R. Ott has been a longtime member of our laboratory. He has officially retired in fall 2005. This has been a big loss for the laboratory in many respects. First, Prof. Ott’s research in low temperature condensed matter physics made him a well known scientist internationally and a recognized leader in condensed matter physics. The world record for the highest critical temperature of a superconductor achieved by his group seems unbroken to this day. His engagement in several other Swiss research and funding institutions, such as the Swiss Science Foundation (Schweizerischer Nationalfonds) and the Paul-Scherrer-Institute, as well as his being chairman of our physics department for the last 4 years demonstrate his broader view of physics and his support for science in general. We are glad that Prof. Ott agreed to stay on as an active researcher for the next two years. We will continue seeking his advice, look forward to more exciting research results from his lab and simply enjoy having him around.

In January 2006 Prof. A. Wallraff started his new research activities in our laboratory. With his unique expertise in superconducting qubits he will extend the research program of the laboratory for solid state physics into a new direction. We extend a warm welcome to him and wish him a lot of success for his research endeavors. He is already involved in several joint projects within the center for ”quantum system for information technology - QSIT” at ETH Zurich as well as in using the FIRST-lab for sample fabrication.

Experimental research at the laboratory for solid state physics benefits from the excellent technical infrastructure provided by the physics department of ETH Zurich. We thank the involved people for their support. We also thank the board of our school for the continuous support of the physics department in general and our laboratory in particular. It is a pleasure to acknowledge the financial support from the Schweizerischer Nationalfonds, the Commission for Technology and Innovation as well as other government and industrial funding sources.

The last year has been a very busy one because of the activities during the World Year of Physics. Members of the institute organized several events in this context, such as a visiting program for primary school children, a night of physics for interested citizens, an Einstein exhibit and a general day at ETH Zurich to reflect about the present and future teaching situation.

I thank S. Gustavsson and Prof. Degiorgi for help in preparing this yearly report.

Zürich, March 2006

Der Vorsteher

Prof. Dr. K. Ensslin
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Chapter 1

Physics of new materials

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1.1 Negative Thermal Expansion in van-der-Waals Bonded Crystals

S. Haas, T. Siegrist (Bell Labs, Murray Hill NJ) and B. Batlogg, in collaboration with C. Besnard and P. Pattison, ETH Lausanne

We have performed complete X-ray structure analyses of tetracene and pentacene in a temperature range from 100–300 K and 100–400 K, respectively. The anisotropic atomic displacement parameters (ADP) were interpreted in terms of librations and translations of the rigid molecules (TLS analysis). Interestingly, we found upon increasing temperature a near-zero thermal expansion along \( a \) in tetracene, and a distinct contraction in pentacene. Upon close inspection of the full expansion tensor and the thermal parameters, we found a consistent explanation assuming that the v-d-W forces tend to minimize the relative shift along the long axis of adjacent molecules. This is further supported by the observation of an unusually large thermal expansion perpendicular to the layers in pentacene, and the indications in our data of a distinctly anharmonic potential for the sliding motion along the long axis. These van der Waals forces leading to negative thermal expansion are expected to be larger in longer molecules, and also to be the driving force behind the well known polymorphism of pentacene.

1.2 SCLC in Organic Single Crystals: Challenges in Reaching the Trap-Free Limit

S. Haas, D. J. Gundlach and B. Batlogg

Densities of trap states of pentacene single crystals have been experimentally determined by the method of temperature-dependent space-charge limited current (TD-SCLC) spectroscopy. With the knowledge of the DOS and by comparing...
the DOS to the corresponding SCLC characteristics, we are able to elucidate challenges associated with reaching the trap-free region, i.e. pushing the quasi Fermi level beyond the mobility edge. The influence of contacts is addressed by the extraction of the activation energy as function of the applied voltage. Surprisingly, we find laminated contacts to be superior compared to thermally evaporated contacts, as shown in figure 1.2. Yet even for laminated contacts, there are some complications at lowest voltage. This is seen as the deviation from the monotonically decreasing dependence of $E_A$ on $V$ at low voltage (shown in the inset). Therefore, the ubiquitously used evaporated Au contacts on organic semiconductors may introduce severe contact effects that are particularly troublesome in 2-point measurements, even if the $I$-$V$ characteristics appears ohmic at low voltage and intrinsically of a SCLC type (cf. black curve in figure 1.2).

### 1.3 Electrical properties of a new rubrene derivative

S. Haas, A. F. Stassen, K. P. Pernstich, G. Schuck, D. J. Gundlach and B. Batlogg

(in collaboration with U. Behrens, H.-J. Kirner and F. Bienewald from Ciba Specialty Chemistry, Basel)

![Figure 1.3: The density of states for high quality rubrene derivative crystals, measured with temperature-dependent SCLC spectroscopy.](image)

The addition of sidegroups to rubrene leaves the in-plane ($a$-$b$) crystal structure almost unchanged, but results in a larger spacing between the naphthacene backbones in $c$-direction. Consequently, the in-plane hole mobility of 12 cm$^2$/Vs measured on single crystal FETs of R1 (see section 1.4) is just as high as in rubrene crystals, as expected from the relationship between molecule packing and charge mobility. The R1 crystals are of high electrical quality, as expressed in the DOS of gap states: A deep-level trap density as low as $10^{15}$ cm$^{-3}$ eV$^{-1}$ has been measured, as well as an exponential band tail with a characteristic energy of 22 meV (c.f. figure 1.3). The bulk mobility, measured in the crystallographic $c$-direction, can be estimated to be of order of $10^{-3} - 10^{-1}$ cm$^2$/Vs.
1.4 Single Crystal Field-Effect Transistors of a New Rubrene Derivative

A. F. Stassen, S. Haas, K. P. Pernstich, G. Schuck, D. J. Gundlach and B. Batlogg

(in collaboration with U. Behrens, H.-J. Kirner and F. Bienewald from Ciba Specialty Chemistry, Basel)

Figure 1.4: The charge mobility along the crystallographic $b$-axis (blue) is four times higher than along the crystallographic $a$-axis (red).

We characterize, in collaboration with Ciba Specialty Chemicals, new organic semiconductor materials that are either functionalized derivatives of known semiconductors, or newly designed molecules.

By investigating chemically closely related materials, the relation between the molecular/crystal structure and the electrical properties can be elucidated.

Transistors are made by placing a single crystal, grown by vapor phase transport, on top of a prefabricated substrate. This way, the almost perfect surface of the single crystal is used for the charge transport. Using single crystals not only results in high quality and reproducible transistors, it also enables to correlate the transport phenomenon to the crystal structure.

In the rubrene derivative R1, charge transport has been measured as good as in rubrene, which is at this moment the organic semiconductor with the highest reported field-effect mobility. The anisotropy in the crystal structure of R1 is reflected in the device mobility: The maximum mobility measured along the $b$-axis is 12 cm$^2$/Vs, while in the same crystal a value of 3 cm$^2$/Vs was measured along the $a$-axis (see figure 1.4). The mobility along the $b$-axis does not strongly depend on the temperature. Upon cooling a typical transistor, the device mobility slightly decreases from 6 cm$^2$/Vs at room temperature to $\sim$2 cm$^2$/Vs at 160 K.

1.5 Determination of the Interface Trap Density of Organic Single Crystal Field-Effect Transistors, and Comparison to the Bulk Trap Density

C. Goldmann, C. Krellner, K. P. Pernstich, S. Haas, D. J. Gundlach, and B. Batlogg

In the quest to optimize the transport properties of organic semiconductors and organic semiconductor devices, it is essential to identify the traps dominating transport at the device interface. In particular, the trap density has to be quantified, especially those traps with capture times of a few hours and less. In our study, the technique of gate bias stress was experimentally developed and applied. Using this method, the density of interface traps in the energy interval defined by the stress bias, which is alternately filled and emptied on timescales given by the stress time, can be extracted. For rubrene single crystal FETs with a typical SiO$_2$ dielectric, this interface trap density was found to be $\sim$ 2 $\cdot$ 10$^{12}$ cm$^{-2}$ at a stress bias of ±50 V. A chemical treatment of the interface with octadecyltrichlorosilane (OTS) molecules forming a self-assembled monolayer reduced this trap density by 50 %. This demonstrates the influence of the interface preparation on charge trapping. In pentacene devices the effect of the interface treatment was of similar magnitude (reduction by 30 %). The measurements also enabled us to quantify the difference in interface trap density between rubrene and pentacene. The trap density was found to be lower by a factor of three for rubrene compared to pentacene on an OTS-treated substrate, and may explain the difference in mobility commonly observed between the two materials.

Further insight could be gained from a qualitative comparison of the interface trap density of the single crystal FETs and the bulk trap density of the single crystals. The latter was determined independently in temperature-dependent
space-charge-limited current (SCLC) measurements. The measured density of charge reversibly trapped and de-trapped at the FET interface was converted into an equivalent volume density; the results for rubrene are shown in Fig. 1.5: The interface trap density of the FETs of order $10^{19}/(\text{cm}^3\text{eV})$ is higher by about three orders of magnitude than the typical bulk trap density. This difference quantitatively highlights the influence on charge trapping of effects intrinsic to the interface, such as polarization. The next step necessary is the further identification and elimination of those traps that are most detrimental for charge transport in organic semiconductor FETs. In this context, bias stress measurements are a convenient tool to assess and quantify the interface trap density.

![Figure 1.5: Comparison of the trap densities (bulk and surface) of freestanding rubrene and pentacene single crystals to the FET interface trap density. The sets of error bars indicate the equivalent densities deduced from the interface trap density of rubrene single crystal FETs with an untreated ("SiO$_2$") and with chemically treated dielectric ("OTS"). These interface densities are several orders of magnitude higher than the bulk trap density determined by SCLC measurements (triangles and squares).](image)

### 1.6 Evidence of Water-related Interface Effects in Pentacene Single Crystal Field-Effect Transistors

C. Goldmann, D. J. Gundlach, and B. Batlogg

![Figure 1.6: (a) Transfer characteristics ($V_{DS} = -10$ V) showing trap formation in a pentacene FET at 297 K for stress times of 0, 15, 35, 60, $\sim107$, 160, 220, and 280 min ($V_{GS} = -60$ V). (b) Saturation value of the discrete trap density at different temperatures. Inset: Time evolution of the trap density at 297 K and 268 K.](image)

In the present work, we discovered during the bias stress measurements an effect that once again underlines the sensitivity of charge transport in FETs to the interface preparation and to effects intrinsic to the interface: In pentacene FETs prepared on untreated SiO$_2$, the generation of discrete trap state with trap densities up to $2 \cdot 10^{12}$ cm$^{-2}$ was observed upon negative bias stress. No trap creation was found in devices in which the dielectric surface had been rendered hydrophobic by an OTS treatment. This is the first strong indication that the process of trap generation is related to a few molecular layers of water adsorbed on the substrate, and thus crucially dependent on the substrate preparation. Fig. 1.6(a) shows the generation of the discrete trap state with increasing stress time at room temperature, Fig. 1.6(b) the maximum trap density that could be created at various temperatures. Interestingly, this trap formation and also the trap relaxation are distinctly different above and below $\sim280$ K. We speculate that the marked difference in trap formation above and below 280 K may be associated with a phase transition in the adsorbed molecular layers of water. While the exact mechanism of trap formation remains to be identified, we note that the timescales observed are typical for diffusion of small ions or molecules in organic single crystals, and suggest a diffusion-limited chemical reaction.
1.7 Novel organic semiconductors for flexible electronics

W. Kalb, A. F. Stassen, K. P. Pernstich, S. Haas and B. Batlogg
(in collaboration with U. Behrens, H.-J. Kirner and F. Bienewald from Ciba Specialty Chemistry, Basel)

In collaboration with Ciba Speciality Chemicals Basel, we are investigating π-electron rich organic materials on their semiconducting properties. The materials are studied in both thin-film transistor structures and single crystal field-effect devices.

We evaluate if a given material has potential as the semiconductor in organic electronics by fabricating field-effect devices. A strong candidate must have a field-effect mobility in thin-films in excess of 0.1 cm²/Vs and should additionally be chemically and physically stable. Furthermore, by comparing different materials, we try to gain information about the extrinsic and intrinsic factors that dominate the charge conduction in the solid state.

![Field-effect mobilities of four new acene-type materials from thin-film and single crystal transistors (left) and output characteristic from a BIF 605 thin-film transistor indicating the high stability of this material (right).](image)

In Figure 1.7 (left) we show field-effect mobilities in four new substituted acene-type materials. The data are derived from thin-film and single-crystal transistor structures. It is remarkable that thin-film and single crystal field-effect mobilities for a given material differ by less than a factor of 15 in all cases despite the large number of grain boundaries inevitably present in thin-films. The output characteristic in Figure 1.7 (right) stems from a thin-film transistor with BIF 605 and shows forward and reverse sweeps of the drain voltage. Interestingly, no current hysteresis is observed for this material, while we observe hysteresis with the benchmark organic semiconductor pentacene in identical transistor structures.

1.8 SiGe modulation doped heterostructures

B. Rössner, H. von Känel, E. Müller
in collaboration with D. Chrastina, G. Isella (Politecnico di Milano)

Silicon-Germanium heterostructures are of high significance for future high-speed electronics, replacing pure silicon. They offer superior performance while being compatible to current processing technology. To make use of pure Germanium as channel material, thick graded buffers are commonly used. The Low Energy Plasma Enhanced Chemical Vapour Deposition (LEPECVD) process, previously developed in this group, is ideally suited for this task, offering growth rates up to 10 nm/s.
Figure 1.8: The measured mobility of backside doped SiGe/Ge heterostructures (symbols) is limited by interface charge scattering for hole densities below $4 \times 10^{11}$ cm$^{-2}$ (dash-dotted line) and by short range scattering at higher carrier density (dashed line). The interface charge density is determined to be $2.4 \times 10^{10}$ cm$^{-2}$. The short range scattering potential has a correlation length $\Lambda = 1.5$ nm and an amplitude of 4 meV.

Figure 1.9: Cross section TEM image of a SiGe/Ge heterostructure: Point defects locally distort the crystal lattice. Their size matches the scattering correlation length determined from mobility measurements. Presumably the substrate temperature was too low during active layer growth.

Precise control over doping profiles is required to fabricate modulation doped devices with the supply layer underneath the channel. Such devices are highly desirable because a top gate can modulate the carrier density in the channel without interference from charging effects in the doping layer which would be present if the doping was above the channel. In addition, due to its larger lattice constant, a Ge channel tends to roughen during deposition, making the upper interface rougher than the lower interface. Thus it is preferable to have the gas closer to the lower interface.

Hall bars fabricated from such material were used to find the dominant scattering mechanisms from the carrier density vs. mobility relation. As can be seen in Fig. 1.8, the mobility rises with carrier density up to about $4 \times 10^{11}$ cm$^{-2}$, then becomes almost independent of carrier density. As charged impurity scattering is characterized by a mobility rising with density, this is a signature of short range scattering. The data are well explained assuming scattering by both interface charges and gaussian correlated scattering. The microscopic origin of the short range scattering can be inferred from a cross section TEM image shown in Fig. 1.9.

The interface charge density of $2.4 \times 10^{10}$ cm$^{-2}$ deduced from the model is remarkably low for SiGe/Ge heterostructures. The current results show potential for an increase of the mobility at densities above $4 \times 10^{11}$ cm$^{-2}$ by an optimized temperature profile during growth.
1.9 Electronic properties of the pyrochlore superconductors RbOs$_2$O$_6$ and KOs$_2$O$_6$

M. Brühwiler, S.M. Kazakov, J. Karpinski, B. Batlogg

The pyrochlores constitute ideal systems to study to what degree itinerant electrons are affected by geometrically frustrated interactions, and what ground states are realized in such systems. This is because the network of the relevant metal atoms consists of corner-sharing tetrahedra. The pyrochlore structure in general is of type $A_2B_2O_6$ with space-group $Fd\bar{3}m$. The recently discovered $\beta$-pyrochlores $AOs_2O_6$ are derived from the parent compound by replacing the O' atoms by Rb or K atoms, leaving the 16d site empty, and occupying the B site by the transition metal cation Os.

A particularly interesting feature of these osmates is the thermal mass enhancement which is due to electron-phonon enhancement and an additional mechanism. We have identified the various contributions to the enhancement reflected in the Sommerfeld coefficient $\gamma$. An interpretation of the additional enhancement in terms of Coulomb correlations requires a parametrization according to $1 + \lambda = (1 + \lambda_{ep})(1 + \lambda_c)$. In this interpretation the overall bandwidth is reduced by a factor of $1 + \lambda_c$ due to electron-electron interactions. The resulting Coulomb enhancement parameter $\lambda_c$ for RbOs$_2$O$_6$ is $\approx 1$ and for KOs$_2$O$_6$ it is $\approx 2$ to 4.

Our data from single crystalline KOs$_2$O$_6$ and polycrystalline RbOs$_2$O$_6$ therefore show that these osmates are particularly interesting transition metal oxides: The effective mass, even after the measured electron-phonon renormalization is taken into account, is two- to fivefold enhanced over the LDA band mass. The renormalization affects those electrons that are paired to form the superconducting condensate.

1.10 MgB$_2$: magnetic and non-magnetic substitutions in single crystals

N. D. Zhigadlo, K. Rogacki, G. Schuck, B. Batlogg, J. Karpinski

Pure and substituted single crystals of the two-gap MgB$_2$ superconductor have been grown at a pressure of 30 kbar at temperature 1800-2000 °C using the cubic anvil technique. The high crystal quality is reflected in a very low residual resistivity and a sharp transition to the superconductivity state, e.g., for nonsubstituted crystals: $\rho_0(40K) \approx 0.5\mu\Omega$cm.
and $\Delta T_c \approx 0.2$ K. Magnetic (Mn, Fe) and non-magnetic (Al, C) ions have been substituted to study their effect on superconductivity and to determine the relative importance of the impurity scattering in and between the $\sigma$ and $\pi$ bands. Mn, Fe and Al substitute Mg, while C substitutes B. Single-phase $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ and $\text{MgB}_2-x\text{C}_x$ crystals were grown for $x=0-0.3$. Al and C dope MgB$_2$ with additional electrons, causing a similar moderate decrease of $T_c$. Substitution of magnetic ions, such as Fe$^{3+}$ and isovalent Mn$^{2+}$ for Mg$^{2+}$ leads to very rapid decrease of $T_c$. Superconductivity is completely suppressed by 2 % of Mn (Fig. 1.11). This reflects the strong coupling between the conduction electrons and the 3d local moments, predominantly of magnetic character, since the non-magnetic ion substitutions, e.g. with Al or C, suppress $T_c$ much less effectively (e.g. 0.5 K / %Al).

![Figure 1.11: Suppression of $T_c$ with Mn substitution for $\text{Mg}_{1-x}\text{Mn}_x\text{B}_2$ single crystals with a sharp transition to the superconducting state. The inset shows $T_c$ versus the lattice parameter $c$.](image1.png)

The normal state magnetic susceptibility has been measured on single crystals with 0.88% Mn substitution, revealing a temperature independent contribution together with an isotropic nearly free ion contribution corresponding to magnetic moment $\approx 1.7 \mu B$ per Mn ion. The magnitude of the magnetic moment uniquely identifies the Mn ions to be divalent, and to be in the low-spin state ($S = 1/2$).

The upper critical field slope $dH_{c2}/dT$ at $T_c$ remains unchanged and equal to 2.2 kOe/K. Fe substitution decreases $T_c$ less rapidly than Mn but much faster than Al and C. Carbon substitution increases the upper critical field twice for $x = 0.05-0.10$, while Al, Fe and Mn substitutions decrease this field. The upper critical field anisotropy decreases with all substitutions, but the temperature dependence of the anisotropy is different, which indicates different scattering rates for different substitutions in the $\pi$ and $\sigma$ bands (Fig. 1.12). Point contact spectroscopy on single crystals substituted with Mn and Al show that the corresponding $\pi$ and $\sigma$ energy gaps exist up to the highest substitution level, while for C substitution, merging of these two energy gaps is observed indicating increased interband scattering (Fig. 1.13).

![Figure 1.12: Upper critical field anisotropy versus reduced temperature for the MgB$_2$ unsubstituted and substituted single crystals.](image2.png)
1.11 Crystal growth, structural studies, and superconducting properties of β-pyrochlore KO₆

S. Kazakov, K. Rogacki, G. Schuck, N. D. Zhigadlo, J. Karpinski

Recently, a new family of β-pyrochlore oxide superconductors AOs₂O₆ (A=Cs, Rb and K) was discovered (S. Yonezawa et al., J.Phys. Condens. Matter 16 (2004 L9)). The structures of α- and β-pyrochlores are shown in Fig. 1.16. For investigations of intrinsic physical and structural properties of this unconventional superconductors single crystals are necessary. Single crystals of KO₆ and RbO₂O₆ have been grown in a sealed quartz ampoule at 600 °C. The crystals are black, have different shapes characteristic for cubic structure (octahedral, cubes) and a size up to about 0.3x0.3x0.3 mm³ (Fig.1.14).

The crystals have metallic conductivity and show a sharp transition to the superconducting state at T_c = 9.65 K for KO₆ and at T_c = 6.5 K for RbO₂O₆. Superconducting properties have been investigated by magnetization measurements performed in a temperature range from 2 to 12 K and in magnetic fields from 0 to 60 kOe (Fig. 1.15). These properties reveal that KO₆ is a type-II superconductor with a rather large Ginzburg-Landau parameter k_{GL} ≈ 70. The crystals show very weak pinning which results in a low irreversibility field.
Single crystal x-ray diffraction studies at room temperature show the presence of Bragg peaks (0kl: k+l ≠ 4n and 00l: l ≠ 4n) that violate symmetry elements of the space group Fd\(^{3}\)m, whereas no Bragg peaks were observed that violate the F-centering. By means of a comparative structure refinement, using both tetragonal and cubic space group symmetries, and through careful examination of possible tetragonal Bragg peak splitting, we conclude that the structure at room temperature is cubic with non-centrosymmetric space group F\(^{4}\)\(^{3}\)m and a = 10.0968(8) \(\AA\). A strong anisotropic character of the potassium channels was observed. Compared to the hypothetical ideal \(\beta\)-pyrochlore lattice (Fd\(^{3}\)m), both osmium tetrahedral and oxygen octahedral network exhibit strong breathing mode-like volume changes (Fig. 1.17).

Figure 1.15: Upper critical field \(H_{c2}\) close to the zero-field superconducting transition temperature \(T_{c} = 9.65\) K for the KO\(_{2}\)O\(_{6}\) crystals. The inset shows a low-field part of the virgin magnetization curve obtained at 5 K for H swept from zero to 5 kOe and then back to zero.

Figure 1.16: The \(\alpha\)-pyrochlore (left) and \(\beta\)-pyrochlore (right) structures shown as a projection along [110]. The ideal \(\beta\)-pyrochlore \(\Box B_2O_6A’ (B = Os; A’ = K, Rb, Cs) structure can be derived from the ideal \(\alpha\)-pyrochlore \(A_2B_2O_6O’\) structure. \(\alpha\)-pyrochlore O’ site is occupied by A’ atoms in \(\beta\)-pyrochlore whereas the \(\alpha\)-pyrochlore A site is empty in \(\beta\)-pyrochlore.

Figure 1.17: Structural details of \(\beta\)-pyrochlore KO\(_{2}\)O\(_{6}\) at room temperature with F\(^{4}\)\(^{3}\)m space group symmetry (middle) compared to Cd\(_2\)Re\(_2\)O\(_{7}\) at 298K (left) and 13 K (right). KO\(_{2}\)O\(_{6}\) (middle): Shown are the O-O interatomic distances in the octahedral network: shown in red increased length and shown in blue decreased length compared to ideal \(\beta\)-pyrochlore structure with Fd\(^{3}\)m space group symmetry. Black spheres: Os; red small spheres O1B; blue small spheres: O1A.
1.12 High-pressure growth of Na osmate single crystals with pyrochlore structure

Z. Bukowski, N.D. Zhigadlo, J. Karpinski

The high-pressure technique has been applied for growing single crystals of Na osmates. The crystallization processes were carried out in a cubic anvil press at 30 kbar with different temperature regimes, starting from mixtures of alkaline metal oxides (peroxides) and OsO$_2$. As a result single crystals of the novel sodium osmate have been obtained. The crystals are black and have octahedral shape with edges up to 1.5 mm long. Structural, thermal and magnetic properties of the crystals have been studied. X-ray diffraction results proved the $\alpha$-pyrochlore structure with $a = 10.170$ Å and the composition of Na$_{1.7}$Os$_2$O$_{6.5}$. This osmate is metallic but not superconducting down to 0.5 K.

1.13 Synthesis of the Ca$_{2-x}$Na$_x$CuO$_2$Cl$_2$ under high pressure

N. D. Zhigadlo, J. Karpinski

The Ca$_{2-x}$Na$_x$CuO$_2$Cl$_2$ compound is a structural analog of a La$_{2-x}$Sr$_x$CuO$_4$ superconductor. Ca$_{2-x}$Na$_x$CuO$_2$Cl$_2$ crystals for $x \approx 0.1$ have been grown under high pressure using the anvil technique. Powders of Ca$_2$CuO$_2$Cl$_2$, NaClO$_4$ (flux, Na source, and oxidizer) and NaCl (flux and Na source) were mixed in dry box and sealed in Pt cylindrical capsule. The sample was compressed at 35 kbar in the high-pressure cubic anvil apparatus. Maximum growth temperature was 1250 °C. As a result black crystals of Ca$_{2-x}$Na$_x$CuO$_2$Cl$_2$ compound of sizes up to 1x1x0.05 mm$^3$ have been grown (Fig. 1.20). By substituting Ca$^{2+}$ with Na$^+$ mobile holes were introduced into the CuO$_2$ planes in the structure. Magnetization measurements show superconductivity at $T_c=13.5$ K in the underdoped state (Fig. 1.19).
In investigations of pure and substituted MgB$_2$, YBa$_2$Cu$_4$O$_8$, Na$_{1-x}$CoO$_2$, KO$_2$O$_6$ and RbOs$_2$O$_6$ in collaboration with other laboratories.

1. Effects of neutron irradiation on superconducting properties of MgB$_2$, Mg(B$_{1-x}$C$_x$)$_2$ and HgBa$_2$CuO$_{4+x}$ single crystals. In collaboration with H. Weber of the Atom Institute in Vienna.

2. Anisotropic properties of MgB$_2$ substituted with Fe, C, Al and Mn by torque magnetometry. In collaboration with R. Puzniak and A. Wisniewski of Institute of Physics of the Polish Academy of Sciences.

3. Scanning Tunneling Spectroscopy of MgB$_2$, (Mg$_{1-x}$Mn$_x$)B$_2$, Mg(B$_{1-x}$C$_x$)$_2$, Mg$_{1-x}$Al$_x$B$_2$ and YBa$_2$Cu$_4$O$_8$ single crystals. In collaboration with Ø. Fischer group, Physics Department University of Geneva.

4. Point-contact spectroscopy of MgB$_2$, (Mg$_{1-x}$Mn$_x$)B$_2$, Mg$_{1-x}$Al$_x$B$_2$ and Mg(B$_{1-x}$C$_x$)$_2$ single crystals. In collaboration with R. Gonnelli group of Politecnico Torino.

5. Isotope effect in the magnetic penetration depth of MgB$_2$. In collaboration with D. Di Castro and H. Keller of Physik Institut Uni Zürich.

6. Magneto-thermopower of single-crystal MgB$_2$ and Mg(B$_{1-x}$C$_x$)$_2$. In collaboration with T. Plackowski of Institute of the Low Temperature and Structure Research in Wroclaw.

7. Haas-van Alphen oscillations in the superconducting state of MgB$_2$ and Mg(B$_{1-x}$C$_x$)$_2$. In collaboration with A. Carrington of University of Bristol.

8. Picosecond dynamics of the superconducting state in MgB$_2$. In collaboration with R. Sobolewski of Rochester University.

9. Magnetic field penetration depth measurements in RbOs$_2$O$_6$ and YBa$_2$Cu$_4$O$_8$. In collaboration with R. Khasanov and H. Keller of Physik Institut of the Uni Zürich.

10. NMR investigations of the charge ordering in Na$_{0.70}$CoO$_2$. In collaboration with J. L. Gavilano and H. R. Ott in our institute.

11. Pressure effects on the transition temperature and the magnetic field penetration depth in the pyrochlore superconductor RbOs$_2$O$_6$. R. Khasanov, H. Keller of Physik Institut Uni Zürich.

12. Inelastic X-ray scattering and X-ray Raman scattering on pure and substituted MgB$_2$. K. Hämäläinen of the Division of X-ray Physics Department of Physical Sciences University of Helsinki.

13. Optical investigations of MgB$_2$, A. Kuzmenko, D. van der Marel, Physics Department University of Geneva.
Chapter 2

Physics of mesoscopic structures, semiconductor nanostructures

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2.1 Spatially resolved transport through a quantum point contact using a scanned probe

A. Pioda, S. Kicin, T. Ihn, and K. Ensslin, in collaboration with M. Reinwald W. Wegscheider, Univ. Regensburg

Quantum point contacts (QPCs) are fundamental building blocks of quantum devices. Their quantized low-temperature conductance observed in very clean samples is a paradigm of the quantum transport of non-interacting electrons. The QPC studied in the experiment was prepared on a GaAs/AlGaAs heterostructure. A two-dimensional electron gas (2DEG) was buried 34 nm below the surface. A mobility $450000 \text{ cm}^2/\text{Vs}$ and an electron density $5 \times 10^{11} \text{ cm}^{-2}$ of the 2DEG were determined at a temperature of 4.2 K. A mesa was fabricated by wet chemical etching in the first processing step. Subsequently the QPC and other quantum structures were defined using local anodic oxidation of the GaAs surface with a room temperature scanning force microscope (SFM). Figure 2.1 shows the resulting topography of the surface. The 2DEG is depleted below the bright oxide lines. The QPC is formed below a 180 nm gap between two orthogonal oxide lines. The transmission of the QPC can be tuned by the in-plane gate. The experiments were performed with a low-temperature SFM in a $^3$He cryostat with a base temperature of 300 mK. The electron temperature was estimated from transport measurements on the neighboring quantum dot structure to be about 550 mK. A tuning fork sensor with an electrochemically sharpened metallic PtIr tip was scanned at constant height above the surface during the experiment. Because it is capacitively coupled to the 2DEG, it can be considered as a scanning gate. From the data we were able to isolate a direct gating effect of the tip measured via the conductance of the point contact. A double tip consisting of two different materials was identified and its induced potential was quantitatively mapped in two dimensions.

2.2 Local investigation of the classical and quantum Hall effect

A. Baumgartner, T. Ihn, and K. Ensslin, in collaboration with D. C. Driscoll, and A. C. Gossard, University of California, Santa Barbara

The transport properties of a 2DEG at high magnetic field are well established and can be understood in the picture of edge states and compressible and incompressible stripes. In the formation of quantum Hall plateaus and in the transition between them, individual potential fluctuations play a major role. Conventional transport experiments can provide informations only about averaged properties and have to be interpreted with statistical models. Direct experimental access to the microscopic mechanisms of the quantum Hall transition can be gained by the use of scanning probe microscopy. Figure 2.2 shows the tip induced changes in the transverse resistance $R_{xy}$ on the left and on the right Hall cross, respectively. If the tip is positioned at the quadrant of corner 1 or 3 (cf. Fig. 2.2(d)) of the Hall cross the measured Hall resistance becomes positive, while it turns negative in the other two quadrants. The AFM-tip couples electrostatically to the sample and changes the local potential, and the electrons are pushed away. One can assume a completely depleted disk beneath the tip with radius $R_{\text{tip}}$. The current density in the Hall bar is then changed. Using electrostatic simulations it is possible to estimate the influence of the tip on the sample potential and the current distribution which then explains the pattern of the Hall effect measured at zero magnetic field. In these simulations it is easy to check the current density flowing into the contact that is closer to the tip position. This reveals that the maxima occur exactly at the position where no more current flows between the nearer corner and the depleted disk. Current densities, on the other hand, come along with a voltage drop, or more intuitively, less current.
means less charge in the contact and thus a less positive Hall voltage. This insight allows one to describe the shape of a butterfly-wing: the tip has the biggest effect if the depleted disk just touches the sample boundary. This explains the maximum running in parallel to the corner in the experiment. The distance then should correspond to the disk radius, as discussed before. The amplitude of the effect decreases far away from the contact entrances, since the tip is no longer relevant for the current entering the voltage probes.

![Figure 2.2](image)

**Figure 2.2:** a) $R_{xy,1}$ and (b) $R_{xy,2}$ in a scanning gate experiment at $B = 0$ and $T = 2.0$ K.

### 2.3 Double quantum dot with integrated charge readout fabricated by layered SFM lithography

M. Sigrist, A. Fuhrer, T. Ihn, and K. Ensslin, in collaboration with D. C. Driscoll, and A. C. Gossard, University of California, Santa Barbara

Coupled semiconductor nanostructures with a high degree of tunability are fabricated using local oxidation with a scanning force microscope. Direct oxidation of the GaAs surface of a Ga[Al]As heterostructure containing a shallow two-dimensional electron gas is combined with the local oxidation of a thin titanium film evaporated on top.

![Figure 2.3](image)

**Figure 2.3:** (a) Conductance through the one left dot measured as a function of the left in-plane gate and two top gate segments . (b) The full charge stability diagram of the double dot system is measured in the QPC detector signal.

Figure 2.3(a) shows a conductance map for transport through one left dot. The tunnel couplings of the other dot are completely pinched off in this regime of voltages. Dark lines correspond to conductance resonances as a function of the two tuning parameters, the left in-plane gate and the top gate segments close to the dot. When the charge detector QPC is tuned into the tunneling regime we are able to detect charging of the individual dots with single electrons.
The charge stability diagram of the double dot system with its characteristic hexagon pattern can be mapped out as a function of two tuning gates [see Fig. 2.3(b)]. The charge detector is closer to the upper dot and therefore shows a stronger signal if this dot is charged. This gives rise to the lines with the smaller slope in Fig. 2.3(b) in agreement with the relative location of this dot to the two sets of tuning gates, respectively. The lines with the larger slope and weaker contrast lie exactly on the conductance maxima presented in Fig. 2.3(a) reflecting the charging of the dot in the lower left. This demonstrates individual control over nanostructured in-plane and top gate electrodes.

2.4 Finite bias charge detection in a quantum dot

R. Schleser, E. Ruh, T. Ihn, K. Ensslin, in collaboration with D. C. Driscoll, and A. C. Gossard, University of California, Santa Barbara

Figure 2.4: (a) Finite bias transport measurement through the dot at a magnetic field of B=0.1 T. Dark regions represent larger positive [blue] or negative [red] differential conductance. (b) Corresponding measurement of the transconductance. Dark regions represent larger values. (c) Single transconductance trace for low bias \( V_{\text{bias}} = 20 \ \mu \text{V}, \) see upper horizontal line in (b)). (d) Single transconductance trace for \( V_{\text{bias}} = -0.2 \ \text{mV} \) (see dashed horizontal line in (b)). (e) Averaged transconductance. Averaging was performed over a set of bias voltages \(-1.6 \ \text{mV} < V_{\text{bias}} < -0.6 \ \text{mV}\). Individual traces were laterally shifted with respect to each other so that the two lines visible in the lower left part of (b) (parallel to the diamond edges in (a)) yield two distinct sharp peaks. The region delimited by dotted lines in (b) marks the range over which averaging took place. Furthermore, a constant and a linear background were subtracted. (f) Averaged transconductance integrated with respect to \( V_{G2} \).

A valuable piece of information about a quantum dot’s characteristics is the knowledge about its coupling to each of the reservoirs. This coupling is determined by the electrostatic barrier forming the constriction and the wave function overlap leading to tunneling. The latter may strongly depend on the quantum state under consideration in the dot, which means that the quantum mechanical tunnel coupling has to be determined for each state individually. We present finite bias measurements of transport through a quantum dot, complemented by simultaneous measurements.
of the conductance of a nearby, electrostatically coupled QPC used as a charge detector. While the latter allows us to
determine the time-averaged charge on the quantum dot even in the non-blockaded regime, the combination of both
methods makes it possible to determine the quantum mechanical coupling of the dot’s energy levels to each of the
two reservoirs. The sample was fabricated using surface probe lithography on a GaAs/Al$_{0.3}$Ga$_{0.7}$As heterostructure,
containing a two-dimensional electron gas (2DEG) 34 nm below the surface as well as a backgate (BG) 1400 nm
below the 2DEG. All measurements were performed in a dilution refrigerator at a base temperature of 80 mK.

Due to the electrostatic coupling of the QPC to the dot, a change in the dot’s charge leads to a modification in the
QPC’s confining potential, resulting in a change of its conductance. The latter was measured by applying a dc voltage
and measuring the resulting current. In the transconductance plot, diagonal lines with the same slope as the diamond
boundaries in the dot conductance plot are observed, marking a change of the dot’s time-averaged charge. The lines
correspond to the alignment of an energy level in the dot with either the source (negative slope) or the drain (positive
slope) reservoir, while their intensity contains information about the magnitude of the change in charge.

Figure 2.4(c) shows the transconductance at low bias. Dips in the signal at the gate voltages of the Coulomb blockade
peaks correspond to the change in electronic charge by one elementary charge each. For a finite bias (Fig. 2.4(d)),
the peaks split, illustrating that the time-averaged charge on the dot changes in steps smaller than the unit charge $e$.
These steps in charge can be directly visualized by integrating over the transconductance (Fig. 2.4(e)-(f)). In order to
improve the precision of the charge determination, the transconductance was averaged over a finite charge range prior
to numerical integration. The large steps marked by dashed horizontal lines in Fig. 2.4(f), corresponding to a change
in charge by one elementary charge each, have almost identical height, as one would expect. Charge rearrangements
during the measurements, one of which is visible at $V_{G2} \approx 235$ mV in Fig. 2.4(a), might contribute to errors in the
numerical integration and thus may lead to small differences in total step height.

To verify the usefulness of the integrated signal as a measure for the mean charge on the dot, we compared the step size
related to a single electronic charge for several Coulomb peaks (see Fig. 2.4(f)) and for different bias ranges between
zero and the height of a Coulomb diamond. The difference in step height did not exceed 10 percent if a sufficient
number of traces was taken into account to reduce the statistical error. Even in the high-bias regime $1.2$ mV < $V_{bias}$ < $1.6$ mV near the border of the Coulomb-blockaded bias range, where the lines in the transconductance seem
to gradually disappear, the integrated transconductance signal gives a good measure of the mean charge, which means
that line broadening compensates for the lower amplitude.

2.5 **Single-hole transistor in p-type GaAs/AlGaAs heterostructures**

Boris Grbic, Renaud Leturcq, and Klaus Ensslin, in collaboration with Dirk Reuter, and Andreas D. Wieck, Univ. of Bochum

Quantum dots implemented in GaAs heterostructures represent promising candidates for the experimental realization
of quantum computation, as well as spintronics devices. However, research based on electronic transport through such
small conducting islands was, so far, exclusively focused on quantum dots defined on n-type GaAs heterostructures.
Here we report on Coulomb blockade (CB) measurements in a single-hole transistor, defined on a p-type carbon doped
GaAs heterostructure.

Nanodevices fabricated on p-type GaAs with conventional split-gate techniques show significant gate instabilities.
This is presumably due to the fact that metallic Schottky barriers on p-type GaAs are more leaky than on n-type
GaAs. Our experience with metallic split-gates on p-type GaAs also shows that they display hysteretic behavior,
making them unsuitable for high precision tuning of the devices. Therefore, we decided to employ another technique,
namely, Atomic Force Microscope (AFM) oxidation lithography to define nanostructures on two-dimensional hole
gases (2DHG).

The crucial point for the implementation of AFM oxidation lithography technique is that the 2DHG is located close to
the sample surface. In case of our heterostructure, the 2DHG resides at the interface 45 nm below the sample surface.
The heterostructure itself consists of 5 nm undoped GaAs cap layer, followed by a 15 nm thick, homogeneously
C-doped layer of AlGaAs, which is separated from the 2DHG by a 25 nm thick, undoped AlGaAs spacer layer.
2.6 Phase coherence in the inelastic cotunneling regime

M. Sigrist, T. Ihn, and K. Ensslin, in collaboration with D. Loss, Univ. Basel, and M. Reinwald W. Wegscheider, Univ. Regensburg

Is electron transport through quantum dots phase coherent? This question roots in the discussion of how to describe it: by incoherent sequential tunneling, or by coherent resonant tunneling? The observation of interference effects have proven that the current through quantum dots (QDs) has phase-coherent contributions. In a quantum dot in the Coulomb blockade regime, the energy gap related to the charging energy becomes larger than $k_B T$ and sequential tunneling transport involving only dot ground states is exponentially suppressed. Up to now, the same suppression was assumed for tunneling involving excited dot states which are spectroscopically accessible at elevated bias where the Coulomb blockade is lifted. In the Coulomb blockade, transport is dominated by cotunneling. Elastic cotunneling, prevalent at low bias voltages, involves virtual tunneling of one electron through the dot via a higher-energy state and leaves the dot in the ground state. Inelastic processes imply correlated tunneling of two electrons, leaving the dot in an excited state with energy $\Delta$ above the ground state. Inelastic cotunneling sets in once the bias energy $eV_{bias} \geq \Delta$.

The sample is based on a GaAlAs heterostructure with a two-dimensional electron gas (2DEG) 34 nm below the surface. It was fabricated by multiple layer local oxidation with a scanning force microscope (SFM).

The resulting AB interferometer has a source and drain opening transmitting at least one mode and being tunable by the top gates sd1 and sd2. One QD is embedded in each arm of the ring. The two dots are tunnel coupled via a quantum point contact (QPC) which constitutes an internal connection between the two branches of the ring.
The conductance was measured in a two-terminal setup at 80 mK electronic temperature. For weak interdot coupling with the dots strongly coupled to the ring the conductance shows an AB period of 22 mT with a visibility (i.e., the ratio of the AB oscillation amplitude and the magnetic field averaged current) up to 0.2 consistent with interference around the entire ring. With negative voltages applied to tqc1-4 the dots can be tuned into the Coulomb blockade regime.

Cross sections through the data are depicted in Fig. 2.7. The phase of the AB oscillations changes by $\pi$ when we cross the inelastic cotunneling onset. At the same time the AB amplitude increases by a factor of 2 and the field averaged (background) conductance increases by a factor of 4. All these changes are well above experimental error. They confirm that at the inelastic cotunneling onset there is a new transport channel taking over in the coherent transport through the second dot. The visibility, in particular, in the elastic cotunneling regime, is exceptionally high indicating that dephasing along the interfering paths is very weak. We interpret the phase change between elastic and inelastic cotunneling observed in Fig. 3(c) as the fingerprint of the excited state in dot 2. The relative phase of the propagating electron between its entrance and exit point contacts depends on the wave function involved. Our measurement shows that there is a phase change of when the state involved in transport changes from the ground to the excited state. The value of $\pi$ is compatible with the phase rigidity expected for a two-terminal measurement and to a possible phase difference of $\pi$ in the tunnel coupling matrix elements of the ground and the excited state in the second dot. The huge numbers found for the visibilities in our experiment are remarkable. We argue that the involved cotunneling processes require a short tunneling time of the order of $h/U \approx 10$ ps ($U$ is half the charging energy) which is short compared to dephasing times of more than 1 ns reported in other experiments. Perhaps even higher order cotunneling processes than those mentioned above as examples can take place.

2.7 Study of the microwave-induced transport through a quantum dot inserted in a 35-GHz loop-gap resonator

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The quantum coherent manipulation of individual spins in quantum dots has attracted much attention as very promising for applications in spin-based quantum information. Several schemes have been proposed to read-out the quantum state of an individual spin via charge transport. In particular, it is predicted that the Rabi oscillations of an electron spin in a quantum dot could produce additional current peaks in the sequential tunneling.

To understand the challenge in this experiment, two facts about quantum dots (QD) are important. First, the Zeeman energy splitting of the unpaired electron spin must be larger than the thermal broadening (3.5kT) of the current peaks observed in sequential tunneling. Given the typical electronic temperature of about 100 mK reached for QD in a standard helium dilution fridge and the g-factor of 0.4 for electrons in GaAs, a Zeeman energy of 125 $\mu$eV (i.e 1.7 K) corresponding to a Larmor frequency of 35 GHz seems reasonable. An important consequence of this is that mw-induced heating must be kept low, a requirement not easily fulfilled in this frequency range for a large mw magnetic field. Second, electrical coupling between the electromagnetic field and the QD must be reduced to a negligible level. This effect produces spurious mw fluctuations of the electrochemical potential of the leads with respect to the electrochemical potential of the QD. As a result, photon-assisted tunneling (PAT) will occur regardless of the spin when the mw electric field is strong.
To excite individual spins in QD within these constrains, our approach is to use loop-gap resonators (LGR). The large amount of mw magnetic energy that can be stored in an LGR has been long recognized at X-band (8-12 GHz). An LGR is an open-shield resonator whose shape, reminiscent of lump circuits, provides a spatial separation between electric and magnetic mw fields on distances significantly shorter than the mw wavelength.

**Figure 2.8:** (a) Sketch of the LGR resonator. The diameter of the smaller loop is 0.45 mm. (b) Nutation of the magnetization of 1 mm3 of -irradiated Ca formate placed in the MW magnetic field on the top of the small loop. (c) AFM–defined quantum dot used in the experiment. Charging energy, level-arm and energy-level spacing, all inferred from the measurement of the Coulomb diamonds (not shown) are: 2 meV, 0.3 and 200 eV respectively. (d) Sketch of the sample inserted into the metallic shield of the LGR. Inset: cut of the GaAs wafer near the optically etched structure supporting the AFM-defined dot.

The LGR, made of silver-coated copper, is pictured in Fig. Fig. 2.8 (a). There is a rectangular window on its side wall. A small elliptic iris couples the LGR to the TE10 mode of a rectangular waveguide. The impedance matching at the entrance plane of the LGR is ensured by a metallic slide-screw tuner located in front of the coupling iris. The typical return loss achieved at base temperature is -20 dB at resonance. The amplitude of the MW magnetic field driving the Rabi oscillations was measured by a standard ESR experiment with a test sample (-irradiated Ca formate). The oscillations of the magnetization of the electron spins shown in Fig. Fig. 2.8 (b) were monitored by integrating the free induction decay (FID) following the MW pulse. The period of these oscillations is a direct determination of the magnetic field strength. Measurements done for different pulse powers provide an efficiency of 0.32 G.mW-5, corresponding to a Rabi frequency of 180 kHz.mW-5 for electrons in GaAs (g-factor 0.4). The quantum dot was fabricated on a AlGaAs-GaAs heterostructure which contains a two-dimensional electron gas (2DEG) 34 nm below the surface. A back gate situated 1.4 µm below the two-dimensional electron gas allows to tune the electron density. An atomic force microscope (AFM) was used to locally oxidize the surface of the semiconductor. As shown in Fig. Fig. 2.8 (c), the 2DEG is depleted below the oxide lines defining a two-terminal quantum dot (dotted circles in Fig. Fig. 2.8 (c)) and a nearby electrostatically coupled quantum-point contact acting as a charge detector (not used here). Charge transport through the dot is controlled via two sets of gates. Dc voltages are applied to the gates G1, G2 to tune the tunnel barriers between the dot and the leads SQD and DQD. All measurements were carried out in a He4-He3 dilution refrigerator with a base temperature of 60 mK and electron temperature of 200 mK without MW.

In order to couple the dot to the MW magnetic field while minimizing the load, the AlGaAs-GaAs wafer was cut as close as possible to the optically etched mesa structure on which the dot is patterned (see Fig. Fig. 2.8 (d)). The positioning of the quantum dot on the top of the small loop of the LGR is checked with an optical microscope.

The set-up used in the present experiment is designed to achieve single electron spin resonance in a quantum dot.
The key element of our approach is a loop-gap resonator, which permits us to reach a relatively high MW magnetic field amplitude in the 35-GHz-range for low MW power. We have shown that the spurious coupling between the quantum dot and the MW electric field, which is detrimental to observing spin resonance, could be fully suppressed by a proper metallic shielding of the GaAs-AlGaAs wafer. Heating caused by the microwave-induced wall currents in the resonator can be strongly reduced in pulse experiments.

2.8 Local oxidation of Ga[Al]As heterostructures with modulated tip-sample voltages

D. Graf, M. Frommenwiler, P. Studerus, T. Ihn, and K. Ensslin, in collaboration with D. C. Driscoll, and A. C. Gossard, University of California, Santa Barbara

![Figure 2.9: SFM images (1.5 μm stripes, lines 1 μm long) with corresponding longitudinal oxide height profile (dashed line marks 10 nm in height) for (a) constant (DC) oxidation voltage and modulated (AC) voltages: (b) f=1 Hz, (c) f=10 Hz, (d) f=100 Hz and (e) f=1 kHz. The lines were written from right to left. For 1 Hz (b) and 10 Hz (c) the pulse scheme of the oxidation voltage is apparent in the oxide height profiles, whereas for higher frequencies (d),(e) the individual sections merge to a continuous line as for the DC case (a). Parameters used: V\text{ox}=-16 V, V\text{res}=4 V, t_{\text{res}}/(t_{\text{res}}+t_{\text{ox}})=50\% (30\% for f=1 kHz). Note that for (c) a different gray scale (top) is used.](image)

In local anodic oxidation a conductive SFM tip acts as the cathode whereas the grounded sample is the anode. A voltage is applied to the tip, while the sample is grounded via a metallic underlay. A thin water film covering the sample surface serves as the electrolyte. The ambient humidity is therefore actively controlled in our setup (42\%). Both, for scanning and writing, the SFM is operated in non-contact mode with a resonance frequency between 315-325 kHz. The feedback loop with amplitude damping as a control parameter is active during the writing process, which is crucial for writing more complex nanodevices consisting of many line sections. Taking the writing velocity of 0.2 μm/s and the base width of the oxide line of about 100 nm into account, a single spot on a line is not oxidized longer than 1 s. This is in contrast to previous experiments on n- and p-doped GaAs, where dots with oxidizing times...
of the order of several seconds were considered. The resonance frequency of the cantilever is two orders of magnitude larger than the frequency of the AC voltage.

In Fig. 2.9 SFM micrographs and height profiles of oxide lines written with DC and AC voltages are presented. With a writing velocity of \( v = 0.2 \, \mu m/s \), the 1 Hz line is just an on/off cycling of shorter DC sections. In the 10 Hz case the individual dots can still be seen if the right contrast is chosen. For further increased frequencies up to 1 kHz consecutive pulses merge to form a continuous line.

Since for 1 and 10 Hz we get a clear segmentation of oxidized and non-oxidized regions [Fig. 2.9 (b),(c)], it is not surprising that the average height taken over all the sections of the line is reduced for low modulation frequencies. However, we stress the fact that for higher frequencies the height of the AC voltage lines reaches almost the one of the constant voltage (DC) lines, despite the fact that the anodizing time has been reduced by 30\% or 50\% since we kept the writing speed constant. The same behavior is observed in the aspect ratio, since the reduction for low frequencies is mostly due to the height averaging. We find that the aspect ratio for \( f = 1 \, kHz \) is enhanced compared to the DC value. On the other hand we can keep the total oxidizing time constant by varying the writing speed. For a duty cycle of 50\% this implies doubling the writing speed in the DC compared to the AC case. We find, that the line height does not vary, but a significant narrowing of the base width can be observed, indicating a reduced lateral carrier diffusion.

We have shown, how improved control over the oxide growth kinetics can be achieved employing a modulated tip-sample voltage. Our results are compatible with the space charge model proposed by Dagata et al. We showed enhanced line aspect ratios and also better reliability and improved reproducibility applying modulated voltage on undoped GaAs. These results motivated us to take advantage of this approach for depleting a shallow two-dimensional electron gas in a Ga[Al]As heterostructure. The electronic stability, the confinement potential and the tunability were investigated in a wire and a point contact geometry. They are found to be qualitatively and quantitatively similar for oxidation with constant and modulated voltage.

2.9 Probing the Kondo density of states in three-terminal quantum rings

R. Leturcq, L. Schmid, and K. Ensslin, in collaboration with Y. Meir, Ben Gurion University, and D. C. Driscoll and A. C. Gossard, University of California, Santa Barbara

The Kondo effect is one of the hallmarks of many-body physics. It was discovered in bulk metals with magnetic impurities providing localized unpaired spins, and observed later in semiconductor quantum dots. The Kondo effect is the coherent coupling of a single unpaired electron spin with a Fermi sea of electron around this single spin. The spins of the surrounding electrons screen this single spin effectively forming a singlet.

In quantum dots the electron reside on a conducting island which is coupled through tunnel contacts with the leads containing Fermi seas. The strength of the coupling is controlled by appropriate gate voltages applied to gates of the device. In this project a novel device has been fabricated, namely a three-terminal quantum ring.

The yellow lines mark lateral barriers being impenetrable for the electrons. Electrons can tunnel from contacts 1 through 3 in and out of the quantum ring.

Figure 2.10: AFM image of the oxide lines defining the three-terminal quantum ring. The three lateral gates marked LG1-LG3 are used to control the conductance of the tunnel contacts. Currents are measured through leads 1,2 and 3. The plunger gates PG control the charge on the quantum ring.
Lateral gates LG1 through LG3 are used to tune the coupling strength, lateral gates named PG are plunger gates which tune the symmetry of the ring and the number of electrons on the device. Whether the single spin residing on the quantum ring has Kondo correlations with the electrons in a given leads depends on the tunneling coupling of the dot to this specific lead. In a three-terminal configuration, which is unique to this experiment, one can determine, which lead has Kondo correlation with the dot and which does not.

By carefully tuning the tunnel couplings different situations can be realized. In the above case one lead termed $\mu_3$ has a large coupling to the dot (thin barrier), and therefore gives to an additional contribution to the density of states in the dot shown by the green peak. The two other leads $\mu_1$ and $\mu_2$ are weakly coupled (thick barrier) and do not provide additional contributions to the density of states. In another situation two leads $\mu_2$ and $\mu_3$ provide Kondo correlations and one lead $\mu_1$ does not.

If there is no bias voltage applied between leads 2 and 3 then there is a single additional contribution to the density of states which results in the blue maximum shown above in the differential conductance trace through the quantum ring. If a bias is applied to the two leads 2 and 3 and they provide two out-of-equilibrium contributions to the density of states which show up as the two maxima of the red curve in the figure above.

This experiment shows several important things: The spin correlations between a quantum system and its leads can be controlled and probed. The additional density of states provided by Kondo correlations can be probed experimentally in and out-of-equilibrium. Three terminal quantum devices allow the determination of intrinsic properties of quantum devices.

The electrical tunability of many parameters of quantum dots makes them a favorable system to study the Kondo effect, in particular out-of-equilibrium, which is an important theoretical issue. Recently, theoretical proposals have shown that quantum dots might be suitable systems for measuring the local density of states (DOS) in the Kondo regime in and out-of-equilibrium, a challenge in scanning tunneling experiments.

For a quantum dot at equilibrium connected to two leads, the screening of the local spin by electrons from both leads gives rise to a peak in the DOS aligned with the chemical potential of the leads, and with a width of order $k_B T_K$, $T_K$ being the Kondo temperature. When a bias larger than $k_B T_K/e$ is applied between the leads a splitting of the enhanced DOS into two peaks aligned with the two chemical potentials of the leads has been predicted theoretically. These peaks will be reduced relative to the equilibrium peak, due to incoherent scattering between the two leads.
Quantum dots are expected to be able to act as single photon sources. When embedded into bulk material, a large amount of the photons are absorbed in the matrix material. In order to avoid absorption in the lateral direction, GaAsP quantum dots embedded in direct band gap GaAsP-material were grown within GaP wires having a diameter of 20 nm and 40 nm, respectively. These wires were grown by low-pressure metal organic phase epitaxy using colloidal gold particles as catalysts. The wires are optically bright and they show several new effects. Their structural properties were investigated by transmission electron microscopy in order to improve the understanding of the origin of their physical properties. High-resolution TEM images reveal that the surface of the wires is not flat but heavily faceted. This is due to twinning as it is typical for the growth of III-V semiconductor material on <111>B substrates. The thickness of the wires corresponds at their top to the diameter of the Au-particles used as catalysts. At the bottom the wires are somewhat thicker, as material is not only built in below the Au-particle but also at the side walls of the wires. The compositional changes within the wire, i.e. GaP to GaAsP, cannot be recognized in the HRTEM images. Energy filtered imaging was used to determine the elemental distribution within the sample. On the elemental P-distribution map a nearly 700 nm long depletion area can be recognized. The P-depletion (corresponding to an As-enrichment) appears to be concentrated in the centre of the wire, while a relatively strong P-signal is present at the wire surface. An EDX-map across a small section of the GaAsP confirms the presence of an inhomogeneous distribution of As and P across the wire. This indicates the presence of a core shell structure having an As-rich core surrounded by a P-rich shell.
Chapter 3

Condensed matter at low temperatures

(https://www.solid.phys.ethz.ch/ott)

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3.1 Low Dimensional Spin Systems

3.1.1 $^{59}$Co-NMR studies of Na$_{0.70}$CoO$_2$

In collaboration with S. M. Kazakov and J. Karpinski

Based on our previous experimental $^{23}$Na- and $^{59}$Co-NMR data and their analysis, we addressed the problem of the character of the Co 3$d$-electron based magnetism of Na$_{0.7}$CoO$_2$. Although our data may consistently be interpreted by assuming that below room temperature the Co 3$d$-electrons are itinerant, this scenario still leaves some open questions. The $^{59}$Co-NMR spectra reveal different Co environments below 300 K. The temperature dependences of the Knight shifts $K(T)$ of two different Co sites, Co1 and Co2 are remarkable in the sense that $K(\chi)$, where $\chi$ is the magnetic susceptibility $\chi = M/H$, display overall non-linear features. Only in two restricted temperature regions ($T > 120$ K and $T < 40$ K) one observes $\Delta K \propto \Delta \chi$, but with different proportionality constants.

A detailed analysis of the spectra allows to identify the spin and orbital parts of the electronic susceptibility, i.e $\chi^{\text{spin}}$ and $\chi^{\text{orb}}$, respectively. With respect to the $^{59}$Co-NMR spectra, the results reveal a dominant role of the local $\chi^{\text{orb}}$. The observed slope changes of $K(\chi)$ are consistent with temperature induced changes of $\chi^{\text{orb}}$ below 100 K. This may be interpreted as evidence for $T$-induced variations in the electronic wave functions of the 3$d$ electrons due, for instance, to an increasing influence of spin-orbit coupling at low temperatures.

Our $T^{-1}_1(T)$ data for the $^{59}$Co nuclei confirm significant variations in the dynamics of this electronic subsystem between 250 and 300 K, as previously suggested [1]. The same data set also allows to distinguish between the spin and orbital part, $T^{-1}_{1,\text{spin}}$ and $T^{-1}_{1,\text{orb}}$, of the spin lattice relaxation rates below 200 K. As expected for Co nuclei in conducting planes, $T^{-1}_{1,\text{orb}} \sim T$. The spin part $T^{-1}_{1,\text{spin}} \propto \sqrt{T}$ is consistent with expectations for a weak antiferromagnet with $T_N = 0$ K. The temperature variation of both the NMR spectra and the spin-lattice relaxation rates indicate that Na$_{0.7}$CoO$_2$ is very close to a magnetic instability at $T = 0$ K. The formation of several inequivalent Co sites at low temperatures and the behaviour of the magnetic susceptibility $\chi(T)$ are presently not understood.


### 3.1.2 Low-temperature Phase Transitions in Na$_{0.5}$CoO$_2$

In collaboration with S. M. Kazakov and J. Karpinski

The compounds Na$_x$CoO$_2$ with $x < 0.5$ exhibit Pauli-type paramagnetism, while for $x > 1$ the magnetic susceptibility $\chi$ exhibits Curie-Weiss type features. The two regions are separated by Na$_{0.5}$CoO$_2$, which exhibits a rapid increase of the electrical resistivity $\rho$ at $T_{MI} = 53$ K [1,2] upon cooling. Preliminary NMR measurements of our group, including the mapping of $^{23}$Na and $^{59}$Co spectra and measuring the $^{23}$Na spin-lattice and spin-spin relaxation rates $T_1^{-1}(T)$ and $T_2^{-1}(T)$, indicated anomalies around $T_X = 88$ K. The metal insulator transition at $T_{MI}$ is reflected by small cusps in $T_1^{-1}(T)$ and $T_2^{-1}(T)$.

In order to clarify the nature of the phases below $T_X$, we made a detailed study of $^{23}$Na NMR spectra in the temperature range between 40 and 300 K. Above $T_X$, a single NMR signal is observed. The temperature and field dependence of the resonance frequency is compatible with a paramagnetic environment. Examples of powder spectra at and below $T_X$ are displayed in Fig. 3.1.2. Below $T_X$, the narrow peak P is strongly reduced and with decreasing temperature we observe two emerging rectangle-shaped signals in the ranges labeled B$_1$-B$_2$ and C$_1$-C$_2$, which we assign to two inequivalent Na sites B and C. It may be seen in the inset of Fig. 3.1.2 that upon cooling, the width of signal B increases abruptly just below $T_X$, saturates towards a constant value and is field independent. The ratio between the widths of signals B and C is 4.2 (temperature and field independent). All these observations provide strong evidence for the ordering of magnetic moments and the subsequent formation of a staggered internal magnetic field in Na$_{0.5}$CoO$_2$ below $T_X$. A more detailed analysis of the temperature variation of the NMR spectra and, in particular, of the quadrupolar features, let us conclude that no significant charge rearrangement accompanies the transitions, neither at $T_X$ nor at $T_{MI}$.

The reported results represent a very important step toward an understanding of the low-temperature properties of Na$_{0.5}$CoO$_2$. A clear result is the onset of an unusual type of magnetic ordering in the metallic phase preceding the metal-insulator transition at $T_{MI}$. It remains a challenge for theoretical investigations, to identify the mechanism(s) which drive the transitions at $T_X$ and $T_{MI}$, respectively.

3.1.3 BaVS₃ at low temperatures

M. Weller, J. Hinderer, J. L. Gavilano, B. Pedrini and H. R. Ott
In collaboration with R. Monnier, B. Delley, PSI, and the group of L. Forro, EPFL Lausanne

BaVS₃ is a compound with quasi one dimensional structural subunits forming chains of V⁴⁺ ions. The compound thus consists of an ensemble of coupled S=1/2 spin chains. The ground state of this system is unknown and the nature of a phase transition at $T = 30$ K, the lowest in a sequence of other transitions at 240 K and 70 K, respectively, is still debated.

Preliminary measurements of the $^{51}$V NMR response and of the magnetic susceptibility revealed the onset of hysteresis effects below 10 K, without any sign of a cooperative phase transition in this temperature range.

In order to clarify the characteristics of the ground state we measured $^{51}$V NMR-spectra of single crystalline BaVS₃ (RRR = 60) at low temperature ($T = 110$ mK) (see figure). In particular we investigated the influence of the orientation of the external magnetic field with respect to the crystal axes in the basal plane of the crystal structure (see figure), and of the thermal history of the sample. The spectra exhibit a very complicated shape and therefore any interpretation of these spectra alone is rather difficult. Nevertheless, some features may clearly be identified. The spectra are very broad; at 67.137 MHz they extend from 4.5 up to 7 T, indicating large electric field gradients at the V sites. Spectra of field-cooled samples are clearly different from those obtained after a zero-field cooling procedure, but the signals cover the same magnetic field interval. For all measured angles of field orientation, the field-cooled samples adopt more spectral weight in the central region of the spectra. Nevertheless, the qualitative shape in the central region is the same for both the field-cooled and the zero-field-cooled situation. This suggests, that for field-cooled samples more V sites experience the same magnetic/electric environment than in the zero-field-cooled specimens.

Figure 3.3: $^{51}$V NMR-spectra at 67.137 MHz of single crystalline BaVS₃ at different orientations of the magnetic field. The c-axis is always perpendicular to the magnetic field orientation, the varied angle is in the ab-plane.
3.2 Magnetism of selected rare-earth compounds

3.2.1 Magnetic order in PrCu$_2$ at 65 K?

B. Pedrini, J. L. Gavilano, M. Weller, and H. R. Ott

The intermetallic compound PrCu$_2$ was previously shown to exhibit an induced structural transition at 7.5 K. It is also known that a cooperative nuclear-electronic magnetic order develops below 50 mK. More recently the results of $\mu$SR-measurements on single-crystalline PrCu$_2$ suggested the occurrence of some magnetic order below 65 K [1].

Aiming at providing additional evidence of the latter claim, we made a series of $^{63}$Cu NMR measurements. Since the material is metallic and, due to the skin effect, the NMR signal originates from nuclei in a thin layer of the surface only, particular care was taken in preparing the single crystalline sample and in mounting it in the NMR coil. These precautions allowed us to record the NMR signal in the temperature range between 20 K and 300 K.

Our first measurements were made in a magnetic field of $H = 7.05$ T, oriented along the a-axis of the orthorhombic structure of the PrCu sample. We identified signals from two different Cu sites with different electric field gradients. In Figure 3.2.1 we display the relative frequency shift of the $^{63}$Cu central line (the same for the two sites) as a function of temperature. The inset shows the temperature dependence of the quadrupolar frequency $\nu_Q$ for the two distinct sites. No anomalies in these data sets may be recognised in the region around 65 K. The same is true for the temperature dependence of the spin-spin relaxation rate $T_2^{-1}$ (not shown), which we also measured between 30 K and 300 K.

Thus our data give no indication to confirm the ordering suggested by the $\mu$SR-experiments, nor for any other type of phase transition around 65 K. Nevertheless, in order to clarify this disagreement we plan further NMR measurements with the magnetic field oriented along either the b- or the c-axes of the PrCu2 sample. In addition to the established data set we plan to measure, if possible, the temperature dependence of the spin-lattice relaxation rate $T_1^{-1}$. Additional NMR experiments at temperatures above and below 7.5 K are intended to probe the structural transition.


3.2.2 NQR studies of CePd$_2$In under hydrostatic pressure

M. Weller, J. Hinderer, J. L. Gavilano, B. Pedrini and H. R. Ott

In collaboration with I. Sheikin, CNRS Grenoble

CePd$_2$ is an intermetallic compound, in which RKKY and Kondo interactions of similar strength compete and thus influence the electronic properties at low temperatures. At ambient pressure, an antiferromagnetic order develops below $T_N = 1.23$ K with a small ordered moment of 0.11$\mu_B$ per Ce-ion. The weak antiferromagnetism and the low Kondo temperature ($T_K \sim 2$ K) make this system interesting for studying its physical properties under pressure [1,2].
3.2. Magnetism of selected rare-earth compounds

We measured $^{115}\text{In-NQR}$ spectra and the corresponding spin-lattice-relaxation rate of $\text{CePd}_2\text{In}$ at 7.5 and 14.6 kbar, respectively [3]. From the spectra we note a pressure induced increase of $T_N$ between ambient pressure and 14.6 kbar, such that $\frac{\Delta T_N}{\Delta p} \approx +16.5 \text{ mK kbar}^{-1}$. A shift in temperature of the characteristic kink in the temperature dependence of the spin-lattice relaxation rate $T_1^{-1}(T)$ at $T_N$ is consistent with this conclusion. The resonance frequency $\nu$ is increasing with pressure, revealing an enhancement of the electrical field gradient at the In site. We assume that by the application of hydrostatic pressure, the RKKY interaction between the Ce 4f moments is gaining in strength with respect to the Kondo screening.

![Figure 3.5: Néel temperature $T_N$ and nuclear quadrupole resonance frequency $\nu$ of CePd$_2$In as a function of pressure.](image)

Figure 3.5: Néel temperature $T_N$ and nuclear quadrupole resonance frequency $\nu$ of CePd$_2$In as a function of pressure.


3.2.3 Magneto-optical evidence of double exchange in a percolating lattice

G. Caimi, A. Perucchi, and L. Degiorgi

In collaboration with V. M. Pereira and A. Castro Neto, Boston University, and H. R. Ott

Because of the potential technological applications, materials exhibiting colossal magnetoresistive (CMR) effects are of high current interest in solid state physics. Europium hexaboride ($\text{EuB}_6$) and the well known manganites, for which the onset of ferromagnetism is accompanied by a dramatic reduction of the electrical resistivity, are primary examples, that have intensively been studied.

We concentrate on the series of cubic $\text{Eu}_{1-x}\text{Ca}_x\text{B}_6$, which displays interesting correlations between magnetic, transport and optical properties. The high temperature electronic transport of $\text{EuB}_6$, a ferromagnet (FM) below $T_C \sim 12$ K, relies on a small effective electron density. The magnetic properties are dominated by the half-filled 4f shell of divalent $\text{Eu}$, which accounts for the measured magnetic moment of $7\mu_B$ per formula-unit. The strong coupling of transport properties to the magnetization was revealed by measurements of the magneto-optical properties, which unveiled a substantial blue shift of the plasma edge in the optical reflectivity with decreasing temperature and increasing magnetic field. The remarkable results correlate with the precipitous drop in the dc resistivity just below $T_C$, and the large negative magnetoresistance observed near $T_C$. The intimate relation between magnetization and electronic conductivity also emerged from experiments on the $\text{Eu}_{1-x}\text{Ca}_x\text{B}_6$ series. The $\text{Ca}$-substitution leads to significant changes of the magnetic and electronic properties. The FM transition temperature decreases with increasing $\text{Ca}$ content and stoichiometric $\text{CaB}_6$ exhibits no magnetic order.

A recently published approach to explain the behavior of $\text{EuB}_6$, as well as offering specific predictions for the electronic properties of the $\text{Eu}_{1-x}\text{Ca}_x\text{B}_6$ series, is based on a double-exchange scenario. This scenario may be regarded as an effective theory for the Kondo lattice problem in the limit of a very small number of carriers. The reduced itinerant carrier concentration places the Fermi level near a magnetization dependent mobility edge, which emerges in the spectral density because of the disordered spin background and/or $\text{Ca}$-doping. A FM metal to insulator crossover is expected as a function of the position of the Fermi level with respect to the mobility edge, which can be tuned by the $\text{Ca}$-content.

The goal of our work is to present and analyze our magneto-optical data of the $\text{Eu}_{1-x}\text{Ca}_x\text{B}_6$ series. Replacing $\text{Eu}$...
by $Ca$ has direct consequences on the electrodynamic response. It influences the distribution of the spectral weight between the metallic (Drude) component and excitations at non-zero energy in the absorption spectrum for different $Ca$-contents at different temperatures and magnetic fields. The Figure displays the variation of the normalized Drude spectral weight (i.e., $\Delta SW^{Drude}/SW^{TOT}$) as a function of $x$, in comparison with the variation of $T_C$ for the $Eu_{1-x}Ca_xB_6$ series. The ranges on the vertical axes were chosen such that $T_C(x = 0)$ coincides with the renormalized changes of the Drude spectral weight, inserting $SW^{TOT}$ for either 0 or 7 T. For both values of $SW^{TOT}$, we obtain the same type of variation with $x$. $\Delta SW^{Drude}$ decreases sharply between $x = 0$ and 0.3, reaching zero at approximately 50% $Ca$-content. The $x$-dependence of $\Delta SW^{Drude}$ reveals the reduction of the maximum of itinerant charge carriers with increasing $x$ in $Eu_{1-x}Ca_xB_6$, previously indicated by results of resistivity Hall effect and optical response measurements. The behaviour of $\Delta SW^{Drude}/SW^{TOT}(x)$ mimics indeed the crossover from a metallic ferromagnet (for $x < 0.4$) to a ferromagnetic Anderson insulator for higher $Ca$ concentration. Our magneto-optical investigations on the $Eu_{1-x}Ca_xB_6$ series provides therefore support for the phase diagram that emerges from the double-exchange model predictions.

Figure 3.6: Dependence of the Curie temperature, $T_C$, on $Ca$-doping, compared to $\Delta SW^{Drude}/SW^{TOT}$, the change of the spectral weight in the Drude component, normalized by the total spectral weight (at 0 or 7 T) encountered in $\sigma_1(\omega)$ up to about 1 eV. The dashed lines for $T_C(x)$ above $x \sim 0.7$ schematically define the interval of $T$ and $Ca$-content, where a cluster spin glass phase was established.
3.3 Superconductivity

3.3.1 Thermal conductivity of the boride superconductors (Mg,Al)B$_2$ and ZrB$_{12}$

A. Sologubenko and H. R. Ott
In collaboration with A. Junod, University of Geneva, and S.M. Kazakov and J. Karpinski

In a previous study we established the influence of C impurities replacing B in MgB$_2$ on the heat transport of this compound [1]. We demonstrated that the substitution of C for B leads to a considerable reduction of the electronic heat transport, while the phonon thermal conductivity is much less sensitive to impurities. The C scattering centers mainly affect the intraband scattering in the $\sigma$-band which predominantly derives from B-based electron states. As is revealed by measurements of the thermal conductivity $\kappa$ as a function of both the temperature and external magnetic fields, the introduction of Al which replaces Mg again mainly affects the electronic heat conduction but, overall, in a less drastic way than C-impurities. While an enhancement of the scattering in the Mg based $\pi$-band is not surprising, we note an equal reduction of the electronic mean free path also in the $\sigma$-band. The influence on the interband scattering seems to be rather weak in both cases.

With the same type of experiments but new single crystals of better quality, we reinvestigated a previously noted and unexpected strong deviation from the Wiedemann-Franz law for MgB$_2$ at low temperatures. From the results of measurements on various different samples we conclude that the previously reported data were not of intrinsic character. In other words, the Wiedemann-Franz law is well obeyed in pure and defect-doped MgB$_2$.

The thermal conductivity $\kappa$, as a function of temperature and external magnetic field, was also measured on single crystalline samples of ZrB$_{12}$, a superconductor with $T_C \approx 6$ K. The aim was to verify a previously claimed temperature induced transition from superconductivity of type I to type II upon cooling. Thermal transport experiments are particularly well suited for this purpose, because the data truly reflect bulk properties of the investigated specimen. Our data confirm the claimed transition at $T^* \approx 4.6$ K but, rather unexpected, we note strongly hysteretic behaviour of $\kappa(H)$ both above and below $T^*$.

3.4 Thermoelectric power of strongly correlated electron systems

3.4.1 Relationship between the thermopower and entropy of strongly correlated electron systems

R. Monnier, in collaboration with V. Zlatic (Institute of Physics, Zagreb, Croatia), J. Freericks (Georgetown University, Washington D.C., USA) and K. Becker (TU Dresden, Germany)

A number of recent experiments report the low-temperature thermopower $\alpha$ and specific heat coefficients $\gamma = C_V / T$ of strongly correlated electron systems. Describing the charge and heat transport in a thermoelectric by transport equations, and assuming that the charge current and the heat current densities are proportional to the number density of the charge carriers, we obtain a simple mean-field relationship between $\alpha$ and the entropy density $S$ of the charge carriers. We discuss corrections to this mean-field formula and use results obtained for the periodic Anderson and the Falicov-Kimball models to explain the concentration (chemical pressure) and temperature dependence of $\alpha/\gamma T$ in $\text{EuCu}_2(\text{Ge}_{1-x}\text{Si}_x)_2$, $\text{CePt}_{1-x}\text{Ni}_x$, and $\text{YbIn}_{1-x}\text{Ag}_x\text{Cu}_4$ intermetallic compounds.

3.4.2 Thermoelectricity of $\text{EuCu}_2(\text{Ge}_{1-x}\text{Si}_x)_2$ intermetallics

R. Monnier, in collaboration with V. Zlatic (Institute of Physics, Zagreb, Croatia) and J. Freericks (Georgetown University, Washington D.C., USA)

The evolution of the thermopower $S(T)$ of $\text{EuCu}_2(\text{Ge}_{1-x}\text{Si}_x)_2$ intermetallics induced by the Si-Ge substitution, is explained by the Kondo scattering of conduction electrons on the Eu ions which fluctuate between the magnetic 2+ and non-magnetic 3+ Hund’s rule ground state configurations. The Si-Ge substitution is equivalent to chemical pressure, which modifies the coupling and the relative occupation of the $f$ and conduction levels.
Crystals growing into a meta stable phase can form complex structures. For snow crystals, it is well known that various morphologies can develop, depending on growth conditions. We study the growth of xenon crystals into undercooled melt. In our experiments xenon is considered as a transparent model substance for metals which allows optical observation of the solidification process in situ in a three-dimensional system. We have observed three morphologies of xenon: dendrites, doublons and seaweed. Transitions between these morphologies might lead to grain refinement, an open question in metallurgy. One goal of the research on structure formation of metals far from equilibrium is a quantitative description of solidification processes. Applications are simulations of the solidification of cast products in automotive industry.

Most work up to now has been focused on the stationary development of the dendrite tips. Theoretical treatments assume a stationary state and a reversible behaviour as it is expected for quasi equilibrium systems. In our studies the development of the whole crystals is taken into account, we study the behaviour of crystals during transient conditions.

A useful means to characterize the influence of convection on solidification is given by the envelope of a dendrite. Three regions can be distinguished: The tip (region I in Fig.1), which can be described by the so called 5/3 law of Brener, region II, where side branches develop according to a Mullins-Sekerka like instability and finally a region of superdendritic growth where coarsening leads to independent side branches. It turns out that convection or other external influences are reflected in the extend how far the three ranges reach. External disturbances do not lead to gradual changes but to sharp transition in the behaviour.

An even more distinct change in growth behaviour is observed if the temperature of the melt is cycled through the melting temperature. We start with a stationary growth. In a first step we increase melt temperature above melting temperature. This leads to a melting back of all fine grained parts of the dendrite and thus to a crystal with a more or less spherical shape. Then we reduce the temperature of the melt below melting temperature and the crystal starts to grow (Part a) and b) in the Fig. 2.). Although the melt temperature decreases, and the growth velocity increases we observe an increase of the tip radius. This is in contradiction to steady state behaviour. Finally the tip becomes unstable and small perturbations emerge out of the sphere. The tip radius decreases abruptly. (Section c) in Fig. 2).

Figure 3.8: Ranges of dendritic behaviour.
I) Ideal shape of a dendrite tip; II) Dendrite tip with side branches; III) Side branches with coarsening and independent growth
Each back melting process provides identical initial conditions and thus it is possible to control the shape of crystals growing into undercooled melt. The discrete jump of the tip radius in these experiments is typical for non-equilibrium experiments which show hysteresis behaviour.

![Figure 3.9: Tip radius and tip velocity during cooling down of the melt. The growth velocity (blue) increases continuously. As a result of a surface instability the spherical shape of the tip breaks down and a small new tip is growing. This leads to a hysteretic behaviour of the tip radius as a function of the temperature of the melt.](image-url)
3.6 Application-oriented thin film research

3.6.1 Epitaxial IV-VI narrow-gap semiconductor layers

M. Arnold, F. Felder, M. Rahim, A.N. Tiwari, and H. Zogg

Narrow gap lead chalcogenide (IV-VI) layers like PbX, Pb\(_{1-x}Sn_x\)X, Pb\(_{1-y}Eu_y\)X and Pb\(_{1-y}Sr_y\)X (X=Te, Se) are investigated for applications and basic research. The band gaps of the active infrared layers are between 0.1 and 0.25 eV (corresponding to wavelengths in the mid-IR range), while higher band gaps are realised with higher y values for the cladding Pb\(_{1-y}Eu_y\)X or Pb\(_{1-y}Sr_y\)X layers. All layers are grown by solid source molecular beam epitaxy (MBE) onto BaF\(_2\) substrates, or onto Si(111)-substrates by employing a CaF\(_2\) buffer layer. The layers are heavily lattice- and (for Si-substrates) thermal-expansion mismatched. Misfit and threading dislocations therefore are present. However, lead-chalcogenides are fault tolerant. Even with threading dislocations densities in the 10\(^7\) cm\(^{-2}\) to 10\(^8\) cm\(^{-2}\) range, optoelectronic devices with reasonable quality are obtained. The inverse minority carrier lifetimes vary proportionally to the density of the threading dislocations, however. Complete heteroepitaxial monolithic two-dimensional Infrared Focal Plane Arrays (2d IR-FPA) on active Si-substrates as well as optically pumped edge-emitting mid-IR lasers, again on Si-substrates, have been realised.

- RCEDs (resonant cavity enhanced detectors) are sensitive within a narrow spectral line only. We fabricated the first such detectors in the mid IR-range which exhibit a single narrow line and high quantum efficiency: As shown in Fig. 3.10, a very thin Pb\(_{1-x}Eu_x\)Se layer forms the photodetector, it is placed inside an optical cavity. The cavity is terminated on one side with an epitaxial non-absorbing Bragg mirror consisting of a stack of Pb\(_{1-x}Eu_x\)Se/BaF\(_2\) pairs. High reflectivities are obtained with a few pairs due to the large difference of the optical indices of PbEuSe and BaF\(_2\). The second mirror consists of Pb which in addition forms the photovoltaic junction.

- VECSELs (vertical external cavity surface emitting lasers) can emit high power and, unlike edge-emitting semiconductor lasers, emit a beam with a very narrow angular spread. We very recently demonstrated the first VECSEL in the mid-IR range emitting at wavelengths above 4 \(\mu\)m. The structures again consist of Bragg mirrors with reflectivities > 99 %, while the active region is just a 1 \(\mu\)m-thick PbSe layer (Fig. 3.11). The exit mirror is curved in order to have a stable resonator. Excitation is done optically using a commercial laser diode emitting at 1.55 \(\mu\)m. The threshold pump intensity has a characteristic temperature \(T_0 = 55\) K, we observed lasing up to 250 K with this first, non-optimized setup.

![Figure 3.10: RCED mid-IR detectors: Schematic structures for two devices, and measured and calculated spectral response for the 4.13 \(\mu\)m peak wavelength device (m order of resonance, all resonances except m=8 are suppressed by absorption).](image-url)
An AFM (atomic force microscope) operating in vacuum at low temperature has been built up. It is used to study dislocations and their movement as well as to obtain I-V characteristics and noise currents of very small photovoltaic sensors (diameter < 5 µm) in-situ. For the latter, we expect a significant change of their noise current if zero, one, two, or more threading dislocations cross the active area of the device.

3.6.2 Thin-film solar cells based on Cu(In,Ga)Se₂ compound semiconductors

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Flexible solar cells

The main challenges associated with flexible solar cells on aluminium or polyimide substrates are growth of CIGS films at low substrate temperatures, since the aluminum foils cannot withstand temperatures higher than 400-500 °C, doping with Na in optimum amounts, and to avoid contamination of the CIGS layer per diffusion from the substrate (Fig. 3.12). Therefore, we have developed growth processes where Na is incorporated after CIGS growth, hence resulting in enhanced crystal quality. The demonstration of the concept using commercially available aluminium foils as substrates led to a conversion efficiency of 6.6 %. This is the first time CIGS solar cells have been successfully grown on aluminium. On commercially available polyimide foils a conversion efficiency of 14.1 % was achieved, which presents a world-record for solar cells grown on polymers.

Figure 3.12: Left: Flexible CIGS solar cells on a 25 cm² aluminium substrate. Right: Secondary ion mass spectrometry (SIMS) depth profile of CIGS/Mo layers in solar cells grown on Al foils at 500 °C show diffusion of Al through the Mo back contact and contamination of CIGS. The Ga-dip/In-hill is due to the growth process and low In-Ga interdiffusion at low temperature.
Microstructural and chemical study of interfaces in CIGS thin-film solar cells

In collaboration with G. Kostorz, ETH Zürich

Interfaces between CIGS and In$_x$S$_y$ buffer layers have been studied by means of bright-field (BF-TEM), high-resolution (HR-TEM) and energy-filtered transmission electron microscopy (EF-TEM), and also by selected-area electron diffraction (SAED), energy-dispersive X-ray spectroscopy (EDX) and scanning electron microscopy (SEM). Efficiencies deteriorate considerably when the In$_x$S$_y$ deposition temperature exceeds about 250 °C, independent of the In$_x$S$_y$ deposition technique. The HR-TEM image in Fig. 3.13 shows an In$_x$S$_y$/CIGS interface where the In$_x$S$_y$ layer was deposited at 300 °C by evaporation of In$_2$S$_3$ powder. A large density of planar defects is visible in the buffer layer. Copious Cu diffusion from CIGS into the buffer was found by EDX. SAED revealed that CuIn$_5$S$_8$ formed instead of In$_x$S$_y$. CuIn$_5$S$_8$ formation has been detected at various In$_x$S$_y$/CIGS interfaces. Its spinel-type crystal structure contains a large number of vacancies and the planar defects shown in Fig. 3.13, which may act as recombination centres at the p-n heterojunction of the solar cell and thus be responsible for the poor photovoltaic performance. Further research topics are the study of Zn(O,S)/CIGS and CdS/CIGS interfaces, as well as the study of MoSe$_2$ for application as buffer layer between CIGS and any metal/semimetal back contact.

Figure 3.13: HR-TEM image of an In$_x$S$_y$/CIGS interface. The In$_x$S$_y$ layer was deposited at 300 °C.

Non-vacuum deposited CIGS

In collaboration with: CREST, Dept. of Electronic and Electrical Eng., University of Loughborough, UK; Institute of Process Engineering, Prof. Dr. S.E. Pratsinis, ETH Zürich; Solaronix S.A., Aubonne; Zentrum für Solarenergie- und Wasserstoffforschung ZSW, Stuttgart, Germany

The cost of processing CIGS solar cells could be dramatically reduced through the use of simple, large-area, non-vacuum deposition techniques. To this end, we are involved in several research projects. One of these projects involves replacing the traditional chemical bath deposited (CBD) CdS buffer layers with ultrasonic spray pyrolysis (USP) deposited In$_2$S$_3$ layers. The USP technique is readily scalable for the coating of large areas and the performance of solar cells processed with the new technique is becoming competitive with our traditional cells, see Fig. 3.14. Another project focuses on developing a novel process for the deposition of CIGS absorber layers. This process currently uses a combination of vacuum and non-vacuum techniques to deposit precursor layers and subsequently convert them into CIS. With further development, the remaining vacuum techniques will be replaced with non-vacuum equivalents. CIS layers prepared by this process have already been used successfully in the production of solar cells.
Figure 3.14: Current–Voltage characteristics comparing two CIGS devices, one for a device processed with the traditional CdS buffer layer, and the other for a device incorporating the new In$_2$S$_3$ buffer layer.
Chapter 4

Magnetism, Electron Spectroscopy

(http://www.microstructure.ethz.ch)

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4.1 Nanoscale Magnetism

Excitations with negative dispersion in a spin vortex


Accoustic membranes resonate in such a way that adding a circular or a diametric node inevitably produces an increase of the vibrational frequency (Chladni law). This phenomenon is a consequence of the positive dispersion of ordinary waves. Negative dispersion – the lowering of the frequency with increasing wavevector – is occasionally measured in light optics for negative index media, special photonics crystals and media with inverted population. In spin systems, negative group velocity was predicted theoretically and excitations with frequencies lower than the uniform precessional mode were interpreted as magneto-static backward modes. We demonstrate by directly imaging the nodal structure that Chladni’s law is violated by spin excitations in small circular ferromagnetic disks (with diameter of the order of micrometer) with a vortex-like ground state configuration. While the precessional frequency indeed increases upon adding circular nodes, it decreases when diametric nodes are built in. We argue that this violation is due to the particular laws governing spin dynamics. This phenomenon can be interpreted as the spin analogon of a photonic left-handed material.

The samples studied are single ferromagnetic permalloy disks with a thickness $d = 15$ nm ranging in radius from 1 to 3 $\mu$m and patterned by e-beam lithography onto a Si substrate. A 500 nm thick Cu micro-coil was prepared around the element using e-beam lithography and electro-plating. It surrounds the sample with an inner diameter of 8 $\mu$m and an outer diameter of 12 $\mu$m. In the ground state the magnetic elements exhibit a flux closure vortex configuration. We perturb this state by launching a short current pulse through the microcoil that produces a magnetic tipping field pulse perpendicular to the sample plane. The maximum strength of the field is less than 50 Oe with a rise time of about 100 ps. This magnetic field pulse exerts a torque onto the local magnetization vector that launches the precessional motion of the elements magnetization. The motion is imaged stroboscopically by means of the magneto-optic Kerr effect as a function of the time delay between application of the tipping pulse and the probe pulse (spatial resolution: $\approx 0.3$ micron). For each magnetic element a movie of the $z$-component of the magnetization vector is recorded at time intervals of 25 – 40 ps. From such a sequence, using a phase sensitive Fourier transform method developed previously in our laboratory, one can reconstruct each individual mode driving the spin motion. The Fourier transform of the time domain data is performed at each location on the disk. Both the amplitude and the phase are recorded. The Fourier transform consists of a sequence of resonances marking the characteristic eigenmodes. The frequencies corresponding to the maxima of the resonance peaks are used for constructing the experimental dispersion curves. The amplitude (top) and the phase (bottom) at resonance are plotted in Fig.1 for the eigenmodes of a disk with $R = 3 \mu$m radius.

Figure 4.1: The spatially resolved spectral weight (top) and phase (down) of the various modes.
One recognizes two types of modes. Some are organized into concentric rings with circular nodes (Fig. 1a-c). The modes in Fig. 1d-e, instead, have one respectively two diametric nodes. The various nodes are easily indentified because at their spatial location the spectral weight is small and because the phase jumps by $\pi$. The modal maps are composed from two half images: the left from the micromagnetic simulation, the right from the experiment. (a) to (c): Axially symmetric modes showing concentric nodes ($n=1,2,3$, $m=0$). (a): 2.80 GHz. (b): 3.91 GHz. (c): 4.49 GHz. (d)-(e): These modes have nodal lines going across the image. (d): 2.08 GHz, one azimuthal node (1,1). (e): 1.69 GHz, two azimuthal nodes (1,2). Notice that across the nodal lines the phase changes by $180^\circ$.

We relate the nodal structure of the modes to a $\vec{k}$ vector and construct experimental dispersion relations. The modal structure observed in Fig. 1 requires the introduction of polar coordinates $r, \varphi$ in the plane of the disk. We define the wavevector $\vec{k} = (k_r, k_\varphi)$ of a spin excitation by introducing an orthogonal set of basis functions suitable for the two-dimensional vortex-like spin configuration. $m_r(r, \varphi) \propto J_1(k_r r) \exp(ik_\varphi \varphi)$. These functions are the analogue for circular geometries of plane waves in cartesian geometries. The $2\pi$ periodicity in $\varphi$ requires $k_\varphi = 0, \pm 1, \pm 2, \ldots$, with the index $m = 0, \pm 1, \pm 2, \ldots$ counting the number of diametric nodes. The boundary conditions at $r = 0$ and at $r = R$ establish a set of possible values for $k_r$: $k_r^n = x_n R$, $x_n$ indicating the zeroes of the Bessel functions $J_1$; $n \in \mathbb{N}$ counts the number of circular antinodes. $n = 1$ corresponds to the state with nodes at $r = R$ and $r = 0$. From Fig. 1 – counting the nodes and their location – one can read out the mode numbers $(n, m)$ of the measured modes. We can now construct the experimental dispersion relations $f$ versus $(n, m=0)$ (Fig. 2, radial modes, left-hand side) and $f$ versus $(n = 1, m)$, (azimuthal modes, Fig. 2, right-hand side). We observe a positive dispersion for the axially symmetric modes $m=0$ and negative dispersion for the modes with $m \neq 0$. In contrast: Chladni’s law for sound in a circular membrane states that $f \propto (m + 2n)^2$. The figure reports the experimental data for disks with different diameters. The dispersion curves show the same behaviour for all disks, their sign being independent of the diameter.

Based on our analytical model, a negative dispersion in magnonic crystals seems to be a quite common phenomenon. Our model shows that introducing spatial nodes does not necessarily mean that the frequency increases, as expected for a large class of phenomena such as ordinary waves but also for quantum-mechanical systems like the hydrogen atom or the harmonic oscillator. In smaller magnetic disks, the exchange interaction should become more important, and it is expected to change the sign of the dispersion. We can envisage the realization of a new type of junction between elements having opposite dispersion, in analogy to the boundary between left-handed and right-handed photonic crystals. If the analogy holds true, a spin wave excitation with $m \neq 0$ should change their helicity when sent across such a junction, thus establishing an elementary spin logic device with subnanosecond switching time.
Micromagnetism in the ultrathin limit

http://www.sciencedirect.com/science/journals

The main experimental observations we would like to address in this paper are the following: *i*) We consider ferromagnetically ordered magnetic element with a thickness in the nanometer range and a lateral size in the micrometer range. When a short magnetic field pulse is applied, spins start a precessional motion which represents an excitation of the equilibrium spin configuration. It has been systematically observed experimentally that the precessional motion proceeds with boundary conditions such that the spins are pinned at lateral boundaries. This remarkable fact has remained unexplained so far. *ii*) We consider about 2 monolayers thick elementary dots made of perpendicularly magnetized fcc Fe epitaxially grown on a Cu(100) single crystal. An extensive study of the domain structure of ultrathin films with \(mm\) lateral size in the same thickness range found the sequence of alternating stripes with ‘↑’ and ‘↓’ magnetization predicted for the infinite monolayer with perpendicular magnetization (stripe width typically in the \(\mu m\) range). We find experimentally that the critical lateral size \(L_c\) of the magnetic element, below which the element becomes single-domain is of the order of the stripe width (i.e. \(\approx \mu m\)). This is in striking contrast to the behaviour in three dimensional elements (i.e. elements extending to similar lengths along all three Euclidean space directions), which become single domain below a lateral size comparable to the domain wall width (i.e. \(\approx nm\)).

The common explanation for these apparently unrelated phenomena is, according to our analysis, a peculiar logarithmic correction of the magnetostatic energy of magnetic elements, produced by the long-range character of the dipolar interaction and by the strictly two-dimensional character of the systems involved. We would like to illustrate this correction in the case of the multi-domain to single domain transition in perpendicularly magnetized magnetic elements. We consider the ultrathin dot as a region of a plane consisting of two domains residing on the right- and left-hand side of a straight wall \(\gamma\) and bounded by the contours \(\gamma_1\) and \(\gamma_2\), see Fig.3.

![Figure 4.3:](image)

We fill the domains with two different spin configurations: one having spin ‘↑’ (i.e. parallel to the film normal) within both domains (Fig.3a) and the second one with spin ‘↑’ on one side and spin ‘↓’ (i.e. antiparallel to the film normal) on the other side of the wall (Fig.3b). Both configurations store some magnetostatic energy arising from the classical dipole-dipole interaction between the magnetic moments. The ‘↑↓’ spin configuration has a lower magnetostatic energy than the ‘↑↑’ one: this favours the formation of domains with opposite magnetization. The magnetostatic energy gain is opposed by the energy cost due to the introduction of a domain wall separating the domains with opposite magnetization. In order for domains to appear, the energy gain must exceed the wall energy. Both energies depend on the size \(L\) of the particle: we will now show that requiring that the two energies be equal establishes a critical lateral size at which a single-domain to multi-domain transition occurs.
The leading contribution to the magnetostatic energy gain \( \Delta E_m(L) \equiv E^{\uparrow\downarrow} - E^{\uparrow\uparrow} \) arises from the self energy of the Amperian current flowing along the segment \( \gamma \). Spreading out the contour \( \gamma \) over a finite domain width \( w \), we obtain, \( \Delta E_m(L) = -2\hat{M}_0^2 \cdot d^2 \cdot L \ln \frac{L}{w} + O(d^2 \cdot L) \) (\( d \) being the thickness of the element). The wall energy is \( E_w(L) = L \cdot d \cdot 2\sqrt{\gamma \Lambda} \) where \( \Gamma \) is the exchange energy between neighbouring spins and \( \Lambda \) is the magnetic anisotropy per unit volume establishing the perpendicular direction as easy magnetization direction. The equation \( E_w(L) + \Delta E_m(L) = 0 \) gives a critical length \( L_c \propto w \cdot e^{(\sqrt{\gamma \Lambda}/\hat{M}_0^2 \cdot d)} \) for the size of the magnetic element, at which a multi-domain to single domain transition should occur. The calculated \( L_c \) has the following properties:

1. In a truly 3D-element extending over length scales \( L \) in three spatial dimensions the magnetostatic energy and the wall energy scale like \( L^3 \) and \( L^2 \), respectively. Thus, the critical size predicted by Kittel’s theory of small particles is of the order of \( \sqrt{\gamma \Lambda}/(M_0^2 \cdot d) \), i.e. it is of the order of the wall width. \( L_c \) for an ultrathin element depends exponentially on this last quantity. Accordingly, the critical size is much larger than the few \( nm \) observed in 3d particles and the wall width of our Fe films (which is below our spatial resolution).

2. Exactly the same exponential function determines the width of stripe domains in continuous infinite perpendicular magnetized ultrathin films: the calculated \( L_c \) is comparable with the stripe width in continuous films.

3. The logarithmic term occurring in the magnetostatic energy gain does not depend on the geometry of the contours \( \gamma_i \) as it arises from the integral along the segment \( \gamma \): the shape of the particle only contributes to the higher order terms. This leads to the result that the particle shape does not affect the argument of the exponential function in \( L_c \) but only contributes to a numerical factor in front of the exponential function.

Notice that there is a large demagnetizing field \((-4\pi\hat{M}_0)\) stored in an ultrathin film with a uniform perpendicular spin configuration \( \hat{M}_0 \). Our results show that this field is not responsible for the formation of domains. Notice also that for an in-plane magnetization configuration we calculate a logarithmic correction in both \( E^{\uparrow\downarrow}(L) \) and \( E^{\uparrow\uparrow}(L) \). However, the leading log cancel out exactly when \( E^{\uparrow\downarrow}(L) \) is subtracted from \( E^{\uparrow\uparrow}(L) \) (provided the wall is exactly parallel to the in-plane magnetization vector). Thus, for an in-plane configuration the magnetostatic energy gain and the wall energy have the same scaling behaviour. In the ultrathin limit there is no value of \( L \) at which it is energetically favourable for domains to penetrate the particle except for the rather unphysical case where \( M_0^2 \cdot d >> \sqrt{\gamma \Lambda} \).

### 4.2 Surface Properties of Quasicrystals

Quasicrystals represent an unusual state of condensed matter, because their atomic structures show orientational long-range order in the absence of periodicity. At an interface where an ordinary crystal and a quasicrystal intersect, the lack of commensurability between the structures leads to misfits on atomic scale resulting in a wealth of novelties. In order to investigate these phenomena, we have used an atomic-beam source and evaporated different materials of a predefined thickness on a well-prepared quasicrystalline surface at a given substrate temperature.

For submonolayer coverage we usually observe epitaxial and continuous growth, because adatoms become trapped at surface vacancies. This will impede surface diffusion and create a seed for further growth. In all heteroepitaxial systems some strain is always present. This strain is first concentrated at those sites where nucleation occurs and changes with film thickness. When the growing film gradually adopts its bulk lattice properties, the strain relaxes. Thus, the strain influences the nucleation kinetics in a layer-dependent way and a competition between the epitaxy-imposed ordering versus the stable bulk phases of the film is encountered in the form of size selection and self orientation of nanocrystals. These crystalline textures on a quasicrystal surface hold promise in a number of important applications.
4.2. Surface Properties of Quasicrystals

Growth of Fe on Al-Pd-Mn

M. Erbudak, Y. Weisskopf

In general, the magnetic anisotropy energy prevents spontaneous changes in magnetization direction. When the anisotropy energy decreases for microscopically small domains, magnetic order is gradually lost. This fact puts a natural limit for miniaturization of magnetic storage devices and calls for strategies to extend the superparamagnetic limit. One obvious solution is to enhance the magnetocrystalline contribution to the anisotropy energy by reducing the crystal-field effects in transition metal $d$-bands and their hybridization with other states. This goal can be achieved by reducing the coordination of atoms by producing still smaller, i.e., nanometric clusters. Therefore, growing a quasiperiodic configuration of self-assembled and self-size selected ferromagnetic nanocrystals would not only be of high interest from a scientific point of view, but also of major significance for potential technological applications.

For this purpose, we have studied the growth of Fe on the fivefold-symmetry surface of the icosahedral quasicrystal Al-Pd-Mn by means of magneto-optical Kerr effect measurements in addition to our usual arsenal of surface-sensitive electron-beam methods. Fe intermixes with the substrate already during the initial stages of growth resulting in an unordered surface region. We have found, that Fe acts as an attractor for Al, which migrates from the underlying quasicrystalline bulk into the adsorbate layer. This process induces a structural modification of the underlying quasicrystalline Al-Pd-Mn region due to Al depletion, thus forming five cubic (110) domains azimuthally rotated by 72° with respect to each other. They are aligned with high-symmetry axes of the icosahedral substrate. Due to the common structural symmetry elements between this cubic Al-Pd-Mn alloy, Fe-rich Al-Fe alloys, and bcc Fe, the growing overlayers gradually adopt this observed orientation of cubic nanoscale domains. Measurements of electron spin polarization in scanning electron microscopy support Kerr results on the in-plane magnetic ordering for films which are thicker than five monolayer equivalents of Fe.

Growth of Si on Al-Pd-Mn

M. Erbudak, J.-N. Longchamp

Quantum confinement of carriers in all three dimensions produces a discrete energy-level spectrum for electrons and holes. This issue is widely known as quantum dots in connection with quantum-information processing and requires fabrication of photonic nano-structures. In practice, common growth techniques do not warrant for a homogeneous size distribution of clusters.

Al and Fe growth modes of nano-structures are largely determined by diffusion and alloying properties of involved elements, respectively. For metals chemically not interacting, energetic considerations, i.e., kinetic control, decide upon the morphology of the growing film. The low-temperature deposition of Al is an example to this growth mode. This argument is in agreement with the fact that metal bonds have no distinct directionality that would influence neighbor interactions in the growing film. The assembly of Si atoms, on the contrary, represents an extremely opposite case of structure formation. By virtue of the symmetry in the electronic $sp^3$ hybrid states, Si crystallizes in diamond structure with the rigid tetrahedral bond angle of 109.5°. Unlike for metals, the bond angle in Si is relatively stiff and lends limited elasticity in fitting the diamond structure to the quasicrystalline substrate during the initial stages of growth. Hence, considerations used to describe metal-atom epitaxy can only be applied to Si with reservation.
We have deposited Si onto the pentagonal surface of Al-Pd-Mn, kept at a mildly elevated temperature of 400 K, using a sufficiently low deposition rate in order to achieve layer-by-layer growth. This temperature is chosen to avoid diffusion of Si into the quasicrystal. For films as thick as 20 AL, we have not detected any structural order using LEED and SEI. The lack of any indication for structural order signals the growth of amorphous or polycrystalline Si without any preferential texture. Experiments at higher substrate temperatures allowing for moderate diffusion of silicon into the quasicrystalline substrate could induce the formation of an intermediate layer, as witnessed in the case of Fe growth on the same surface, and facilitate crystalline growth. These efforts are under way.
Chapter 5

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H.M. Singer and J.H. Bilgram
Three-Dimensional Single Crystal Morphologies of Diffusion Limited Growth in Experiments and Phase Field Simulations

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A. Vindigni, A. Rettori, L. Bogani, A. Caneschi, D. Gatteschi, R. Sessoli, and M.A. Novak
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P. Wachter and B. Bucher
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Y. Weisskopf, R. Lüscher, and M. Erbudak
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H. Wilhelm, D. Jaccard, V. Zlatic, R. Monnier, B. Delley and B. Coqblin
High-pressure transport properties of CeRu$_2$Ge$_2$

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Onset and amplitude of sidebranches in three dimensional growth of xenon dendrites

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Time Resolved Magnetization Dynamics of ultrathin Fe(001)-films: Spin-Pumping and Two-Magnon Scattering

Magnetic properties of superconducting HgBa$_2$CuO$_{4+x}$ single crystals in the overdoped state before and after particle irradiation

V. Zlatic and R. Monnier
Theory of the thermoelectricity of intermetallic compounds with Ce or Yb ions
Chapter 6

Talks

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* Batlogg B.
Superconductivity in pyrochlore-structured osmates
Workshop on Novel Materials and Superconductors, Planeralm, Austria, 13.02.2005

* Batlogg B.
Trap spectroscopy in crystalline organic semiconductors
Summer School on Organic Electronic Materials and Devices, Alghero/Sardegna, Italy, 13.06.2005

Batlogg B.
Enhanced low-energy excitations in Na$_x$CoO$_2$
Int. Conference on Strongly Correlated Electron Systems, Vienna, Austria, 29.07.2005

* Batlogg B.
New and old questions in oxide superconductors
Paul Scherrer Institut (PSI), Villigen, Switzerland, 18.10.2005

Batlogg B.
Na$_x$CoO$_2$: Strong mass enhancement on the triangular lattice
ESF Workshop on Highly Frustrated Magnetism, La Londe, France, 07.11.2005

* Bilgram J.
Physik des Wachstums von Xenonkristallen einfach komplex - Bildbäume und Baumbilder in der Wissenschaft
Ausstellung im Museum für Gestaltung, Zürich, Switzerland, 30.04. - 04.09.2005

* Bilgram J.
Nonlinearities, Chaos and Fractals, 1. Nonlinear Processes
Seminar, Institute of Low Temperature Sciences, Sapporo, Japan, 09.09.2005

* Bilgram J.
Nonlinearities, Chaos and Fractals, 2. Fractals
Seminar, Institute of Low Temperature Sciences, Hokkaido University. Sapporo, Japan, 12.09.2005

* Bilgram J.
Nonlinearities, Chaos and Fractals, 3. Wavelets
Seminar, Institute of Low Temperature Sciences, Hokkaido University. Sapporo, Japan, 13.09.2005
* Bilgram J.
Complex Structures: A Symbiosis of Experiments and Numerical Studies
Colloquium, Institute of Low Temperature Sciences, Hokkaido University, Sapporo, Japan, 15.09.2005

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Complex Structures: A Symbiosis of Experiments and Numerical Studies
The Physical Society of Japan (JPS), Annual meeting, Kyoto, Japan, 20.09.2005

* Bilgram J.
Controlling of Complex Structures
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Komplexe Strukturen
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Büchwiler M.
Large Mass Enhancement in the Metallic Pyrochlore RbOs$_2$O$_6$
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Büchwiler M.
Strongly Correlated Pyrochlore Superconductors KO$_2$O$_6$ and RbOs$_2$O$_6$
MaNEP 2005; Les Diablerets, Switzerland, 26.09.2005

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Itinerant Correlated Electrons on Triangular Lattices
European Science Foundation Highly Frustrated Magnetism Workshop; Agelonde, La Londe des Maures, France, 08.11.2005

Buess M.
Excitations with negative dispersion in a spin vortex,
DPG März-Tagung, Berlin, 4-9.03.2005

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Micromagnetic Dissipation, Dispersion and Mode Conversion in thin permalloy platelets,
DPG März-Tagung, Berlin, 4-9.03.2005

Caimi G.
Magnetically driven structural transition in Cu$_3$TeO$_6$

* Degiorgi L.
Light and Matter
Physics Colloquium at University of Lipsia, Lipsia, Germany, 4.1.2005

Degiorgi L.
Charge density wave gap in ZrTe$_3$
March Meeting of the American Physical Society, Los Angeles, U.S.A., 21-25.3.2005
Degiorgi L.
Optical investigations on Na$_{0.7}$CoO$_2$
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* Degiorgi L.
Electrodynamic response of strongly correlated systems
Physics Colloquium at University of Neuchatel, Neuchatel, Switzerland, 18.4.2005

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Einstein: Annus Mirabilis
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Magneto-optical investigation on novel superconductors: MgB$_2$ and Na$_{0.7}$CoO$_2$

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Ellenberger C.
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Niels Bohr Summer Institute on Transport in mesoscopic and single-molecule systems, Copenhagen, Denmark, 23.08.2005,

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Departament de Fisica, Universitat de les Illes Balears, Palma de Mallorca, Spain, 11.11.2005

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