Ultrafast dephasing of continuum transitions in bulk semiconductors

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We have studied the coherent emission from bulk semiconductors by spectrally resolved and spectrally integrated four-wave mixing (FWM) with broadband 16-fs pulses in the carrier density range from $1\times10^{16}\,\mathrm{cm}^{-3}$ to $6\times10^{17}\,\mathrm{cm}^{-3}$. In GaAs, only continuum transitions are excited, while both excitonic and continuum transitions are excited in $Al_{0.06}Ga_{0.94}As$. The decay of the FWM signal for positive time delays unambiguously reflects the dephasing of the continuum, irrespective of the excitation of excitonic transitions. We find identical ultrafast sub-20-fs decay times of the coherent emission from continuum transitions in GaAs and $Al_{0.06}Ga_{0.94}As$, independent of the excess excitation energy. These decay times depend only weakly on the carrier density for densities below $10^{17}\,\mathrm{cm}^{-3}$. The continuum dephasing is analyzed considering carrier-carrier and carrier-LO-phonon scattering in a relaxation rate approach. The excitonic transitions in $Al_{0.06}Ga_{0.94}As$ dominate the FWM signal at negative time delays. [S0163-1829(99)00824-3]

In pioneering works, dephasing in semiconductors was investigated with broadband sub-10-fs pulses, simultaneously exciting transitions at the band edge and continuum transitions high in the band.[1,2] In bulk GaAs, decay times of the coherent emission were obtained from spectrally integrated four-wave-mixing (FWM) experiments in the high-carrier density regime for $N>1\times10^{17}\,\mathrm{cm}^{-3}$.[1] The decay was attributed to dephasing of continuum transitions, neglecting possible contributions from the band edge since the major part of the excitation laser spectrum overlapped with the continuum.[1] The dephasing times of only a few tens of femtoseconds were explained by pure carrier-carrier scattering due to screened Coulomb interaction.[1] LO phonon scattering was neglected.[1]

Later it was found that the excitonic contribution to the FWM signal was much stronger than the continuum contribution.[3] Therefore, it was questioned whether the dephasing times measured without spectral resolution in Refs. 1 and 2 reflected only the dephasing of the continuum.[3] Spectrally resolved FWM experiments with 100-fs pulses involving continuum transitions did not provide good enough time resolution to clearly resolve the dephasing of continuum transitions.[4,5] In more recent FWM experiments with 14-fs pulses centered at the band edge, the spectrum of the coherent emission had its peak at the band edge and only at high densities well above $10^{17}\,\mathrm{cm}^{-3}$ the dephasing of continuum transitions could be analyzed.[6] The decay of the coherent FWM emission from continuum transitions on the 10-fs time scale has recently attracted considerable interest by theory due to its quantum kinetics aspects. Theory has concentrated on either the quantum kinetics of Coulomb scattering[7,8,9] or LO phonon scattering.[10,11]

In this paper, we study the decay of the coherent FWM emission from continuum transitions in bulk semiconductors in dependence of the carrier density and the excess excitation energy with respect to the band edge. Spectrally resolved FWM allows us to unambiguously relate the decay of the coherent emission to either excitonic or continuum transitions. We find that the decay of the FWM signal at positive time delays is due to continuum transitions even if excitonic transitions are excited together with the continuum in our experiments. Continuum dephasing is studied at carrier densities more than one order of magnitude lower than in Ref. 1 and ultrafast decay times well below 20 fs are obtained even at the lowest density of $1\times10^{16}\,\mathrm{cm}^{-3}$. While our data reproduces the high-density results of Ref. 1, we obtain a much weaker density dependence of the continuum dephasing for lower carrier densities due to the increased importance of density-independent LO phonon scattering. The dependence of the continuum dephasing on the excess energy is negligible in the density range investigated. It is shown that Coulomb and LO phonon scattering need to be considered to model continuum dephasing at lower densities. An approximate model is obtained by a simple relaxation rate approach. We will discuss the impact of our experimental results with respect to quantum kinetics theory.

We have performed FWM experiments with 16-fs pulses from a Ti:sapphire laser in the standard two-beam configuration. The laser spectra are centered at 1.54 eV and have a full width at half maximum of 75 meV. The two excitation pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are parallel polarized. The FWM signal, in direction $2\mathbf{k}_2-\mathbf{k}_1$, is detected either spectrally integrated versus the time delay Δt between the excitation laser pulses or spectrally resolved at fixed time delays Δt. We have investigated a GaAs sample and an $Al_xGa_{1-x}As$ sample with $x=6\%$ aluminum content, both having a thickness of 200 nm. To allow for transmission experiments, the samples were glued on sapphire disks and the GaAs substrate was removed by wet etching. The samples are antireflection coated at the front side. All experiments have been performed at room temperature.

The use of the GaAs and the $Al_{0.06}Ga_{0.94}As$ sample allows us to vary the excitation conditions even though the laser spectrum is the same in all experiments. This is illustrated in the insets of Fig. 1. In the GaAs sample, the peak of the excitation laser spectrum lies 120 meV above the band edge. The band edge is not excited and excitonic transitions are not expected to contribute to the FWM signal. In order to investigate the influence of excitonic transitions on the FWM signal, the $Al_{0.06}Ga_{0.94}As$ sample is used whose band edge overlaps with the low-energy flank of the excitation laser spectrum. The peak of the excitation laser spectrum lies 40

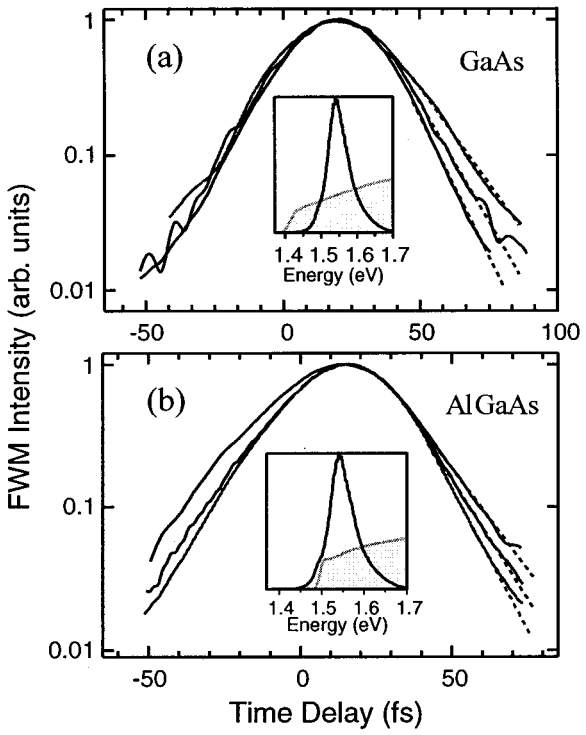

FIG. 1. Spectrally integrated four-wave-mixing signals (solid lines) from the GaAs sample (a) and the $Al_{0.06}Ga_{0.94}As$ sample (b) vs time delay at room temperature. Carrier densities $N=6\times10^{17}$ (steepest slope), $N=7\times10^{16}$, and $N=2\times10^{16}\,cm^{-3}$ in (a) and (b). Dashed lines: exponential fits. Insets: Linear absorption spectra at room temperature (thin line) and excitation pulse spectrum (thick line).

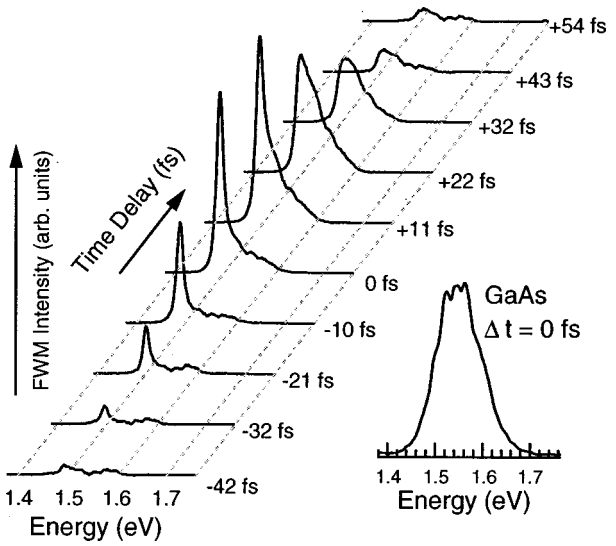

FIG. 2. Spectrally resolved four-wave-mixing signal from the $Al_{0.06}Ga_{0.94}As$ sample for different time delays at room temperature, carrier density $N=2\times10^{16}\,cm^{-3}$. Inset: Spectrally resolved four-wave-mixing signal from the GaAs sample for the time delay $\Delta t=0$ fs at room temperature, carrier density $N=4\times10^{16}\,cm^{-3}$.

meV above the band edge of this sample. It is important to note that the excess excitation energy with respect to the band edge is increased by 80 meV in the GaAs sample as compared to the $Al_xGa_{1-x}As$ sample.

Figure 1(a) shows the spectrally integrated FWM signal from the GaAs sample versus time delay Δt for different carrier densities from 2×10^{16} to $6\times10^{17}\,cm^{-3}$. A carrier density dependent decay is observed for positive time delays, which becomes slower for decreasing carrier density. This proves that at least the decays of the FWM signals for carrier densities lower than the maximum density are clearly resolved and reflect the dephasing of the coherent emission. The dashed lines represent fits to an exponential function $\propto\exp(-\Delta t/\tau)$ with decay times τ, which vary from 15.4 to 10.4 fs for carrier densities between 2×10^{16} and $6\times10^{17}\,cm^{-3}$. The fastest decay time of 10.4 fs is very close to the time resolution of the experiment. The rise of the FWM signal is determined by the pulse width for all carrier densities.

In contrast, the FWM signals from the $Al_{0.06}Ga_{0.94}As$ sample, plotted in Fig. 1(b) for the same carrier densities as in Fig. 1(a), show a density-dependent rise at negative time delays. The comparison between the results from the GaAs and the $Al_{0.06}Ga_{0.94}As$ sample suggests that the density dependent rise of the FWM signal from the $Al_{0.06}Ga_{0.94}As$ sample is due to the excitonic transitions. The rise becomes slower with decreasing carrier density. The same density dependence is found for the decays at positive time delays, which can be fitted to exponential functions $\propto\exp(-\Delta t/\tau)$ with time constants τ varying from 15.3 to 11.0 fs for carrier densities from 2×10^{16} to $6\times10^{17}\,cm^{-3}$ [dashed lines in Fig. 1(b)].

Spectrally resolved FWM was performed to analyze the contributions to the coherent emission. Figure 2 shows the results for time delays from -42 to $+54$ fs for the simultaneous excitation of continuum and excitonic transitions in the $Al_{0.06}Ga_{0.94}As$ sample. The carrier density is $N=2\times10^{16}\,cm^{-3}$, corresponding to the curve with the slowest decay in Fig. 1(b). An excitonic contribution to the FWM signal is clearly visible at the band edge and dominates the FWM power spectrum for time delays from -32 to $+11$ fs. For time delays $\Delta t>11$ fs the FWM power spectrum broadens and the excitonic contribution is no longer dominant. The excitonic contribution vanishes for time delays $\Delta t>22$ fs. The data in Fig. 2 clearly show that the excitonic transitions give rise to the FWM signal at negative time delays, but yield only a negligible contribution at positive time delays. Similar results have been obtained in FWM experiments with 100-fs pulses exciting both excitonic and continuum transitions.[5,12] These results have been explained by excitonic polarization scattering,[13,14] which, at negative time delays, gives rise to an enhancement of the homogeneously broadened excitonic contribution to the FWM signal relative to the inhomogeneously broadened continuum contribution since it is only effective for homogeneous transitions.[5,12,15] At positive time delays, phase space-filling results in FWM emission from both the continuum and the excitonic transitions, leading to a relative decrease of the excitonic contribution.[5,12,15]

As expected, there is no excitonic contribution in the coherent FWM emission from the GaAs sample, as shown in the inset of Fig. 2 for the time delay $\Delta t=0$ fs and the carrier density $N=4\times10^{16}\,cm^{-3}$. The FWM power spectra from the GaAs sample hardly change their shape with varying time

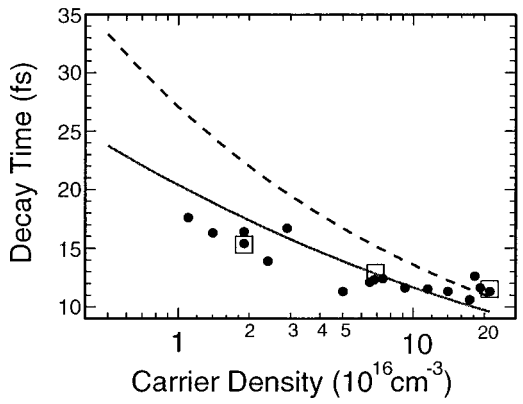

FIG. 3. Decay times of the spectrally integrated four-wave-mixing signals from the GaAs sample (filled circles) and the $Al_{0.06}Ga_{0.94}As$ sample (open squares) vs carrier density at room temperature. Dashed line: extrapolation of the high-density results from Ref. 1. Solid line: extrapolation of the high-density results from Ref. 1 with additional electron-LO-phonon scattering with an electron-LO-phonon scattering time of 165 fs (Ref. 20).

delay and carrier density. We note that, with increasing carrier density, the excitonic contribution to the coherent emission from the $Al_{0.06}Ga_{0.94}As$ sample decreases compared to the continuum contribution (data not shown), as observed previously.[4,15] Therefore, we conclude that the decay of the spectrally integrated FWM signal for positive time delays unambiguously reflects the dephasing of the continuum transitions in the whole density range from 1×10^{16} to $6\times10^{17}\,\mathrm{cm}^{-3}$, irrespective of the excitation of excitonic transitions.

We will now discuss the dephasing of the continuum in more detail. Figure 3 shows the decay times τ of the coherent emission as a function of the carrier density for the GaAs (filled circles) and the $Al_{0.06}Ga_{0.94}As$ (open squares) sample. These decay times have been extracted from exponential fits at positive time delays to a large set of FWM traces, and unambiguously reflect the dephasing of the continuum. The data for the highest density are not included in Fig. 3 since they are close to the time resolution of the experiment. As possible dephasing mechanisms, Coulomb scattering and LO-phonon interaction should be considered. Scattering with acoustic phonons is slower than LO-phonon scattering[3] and, therefore, is not expected to be important in FWM experiments on the 10-fs time scale. We note that even the energetically highest electrons excited by our laser pulses are too low in energy for resonant intervalley scattering[16] to happen in the GaAs and the $Al_{0.06}Ga_{0.94}As$ sample. Consequently, intervalley scattering does not have to be considered in our experiments. Moreover, alloy scattering in $Al_{0.06}Ga_{0.94}As$ is known to make contributions of less than 1 meV to the homogeneous broadening.[17] Therefore, it is negligible on the 10-fs time scale.

Figure 3 shows that the density dependence of the decay times is very weak for both samples. Decreasing the density by one order of magnitude from 2×10^{17} to $2\times10^{16}\,\mathrm{cm}^{-3}$ results in an increase of the decay times of less than a factor 1.4. We will now compare these results to previous work on bulk GaAs, performed in the high-density regime from 2×10^{17} to $7\times10^{18}\,\mathrm{cm}^{-3}$, and reported in Ref. 1. First, we note that our measurement at $N=2\times10^{17}\,\mathrm{cm}^{-3}$ reproduces the result obtained in Ref. 1 at this density. This confirms that (i) the time resolution in our experiments is sufficient to determine the decay time at densities $N\leqslant2\times10^{17}\,\mathrm{cm}^{-3}$, and (ii) that the continuum dephasing decay times in Ref. 1 are not affected by the excitation of the band edge, as expected from our spectral analysis in Fig. 2. The measurements in the high-density regime have shown that the decay time is given by $\tau_{\mathrm{HD}}(\mathrm{fs})=6.8[N(10^{18}\,\mathrm{cm}^{-3})]^{-0.3}$, for densities N from 2×10^{17} to $7\times10^{18}\,\mathrm{cm}^{-3}$.[1] This relation has been explained accounting for carrier-carrier scattering due to strongly screened Coulomb interaction with an effective screening length given by the average nearest-neighbor distance between carriers.[1,2] This model predicts $\tau_{\mathrm{HD}}\propto N^{-1/3}$, in agreement with the high-density data in Ref. 1. Carrier LO-phonon interaction was neglected in the high-density regime.[1] The dashed line in Fig. 3(a) shows the relation $\tau_{\mathrm{HD}}(\mathrm{fs})=6.8[N(10^{18}\,\mathrm{cm}^{-3})]^{-0.3}$ extrapolated to lower densities. Our data in Fig. 3 demonstrate that significantly shorter decay times of the continuum dephasing are obtained below $10^{17}\,\mathrm{cm}^{-3}$ than predicted by the extrapolation of the high-density results. This is equivalent to a weaker density dependence of the decay below $10^{17}\,\mathrm{cm}^{-3}$, i.e., to a weaker increase of the decay times for decreasing density. These results indicate that, at carrier densities below $10^{17}\,\mathrm{cm}^{-3}$, another process contributes to the dephasing besides Coulomb scattering. This process should have a weak-density dependence, such as LO-phonon scattering.[18,19]

Electron-LO-phonon scattering times of 165 and 132 fs have been experimentally determined in Refs. 20 and 21, respectively. Theoretically, an electron-LO-phonon scattering time of 220 fs was predicted.[22] In the following, we will use an electron-LO-phonon scattering time τ_{LO} of 165 fs (Ref. 20) independent of the density, which is justified for $N\leqslant10^{17}\,\mathrm{cm}^{-3}$.[18,19] Adding the electron-LO-phonon scattering rate $1/\tau_{\mathrm{LO}}$ to the high-density FWM decay rate $1/\tau_{\mathrm{HD}}$,[1] which neglects electron-LO-phonon scattering, we obtain for the FWM decay time at lower densities $\tau=(1/\tau_{\mathrm{HD}}+2/\tau_{\mathrm{LO}})^{-1}$. Electron-LO-phonon scattering is accounted for as an energy relaxation process[23] and the semiconductor continuum is treated as an inhomogeneously broadened system[24] to derive this relation. τ is plotted as solid line in Fig. 3 and reasonable agreement with the experimental results is found. To the best of our knowledge, the data of Fig. 3 represent the first experimental proof that the strong-screening model for carrier-carrier scattering of Ref. 1 together with the electron-LO-phonon scattering time can approximately describe the dephasing time of the semiconductor continuum at lower densities.

This is somewhat surprising because a quantum kinetics description of the Coulomb scattering[7,8,9] seems more appropriate during the built up of screening on the 10-fs time scale than the strong-screening model.[1] Moreover, on these time scale, a quantum kinetics description of electron-LO-phonon scattering is required.[10,11] Coulomb quantum kinetics predicts a decay of the spectrally integrated FWM signal $\propto\exp(-\Delta t^3/\Theta^3)$ with $\Theta\propto N^{-1/3}$,[7,8] also obtained from a quasiclassical theory.[25] We have fitted the experimental curves to the above relation although this relation did not reproduce the experimental results as well as the exponential fits shown in Fig. 1. A considerably weaker density dependence of the

time constant Θ was found than predicted by Coulomb quantum kinetics. This again indicates that, for densities below $10^{17}\,\mathrm{cm}^{-3}$, both LO phonon and Coulomb interaction have to be considered to describe the dephasing of semiconductor continuum transitions. To our knowledge, a quantum kinetics description of the loss of coherence of semiconductor continuum transitions has not yet been worked out accounting for both Coulomb and LO-phonon scattering. It needs to be determined in which respect quantum kinetics can improve the description of the experimental results.

We will now comment on the energy dependence of the continuum dephasing. Figure 3 shows that almost identical decay times of the continuum dephasing are obtained in the GaAs and the $Al_xGa_{1-x}As$ sample for all densities. Recalling that the excess energy varies by 80 meV between the samples, this demonstrates that the dephasing of the continuum is almost independent of the excess energy for densities below $10^{17}\,\mathrm{cm}^{-3}$. This is plausible given that Coulomb scattering in the quantum kinetics regime is only weakly wave vector and energy dependent.[6,7] We note that the negligible energy dependence justifies the use of a single time constant to describe the decay of the coherent emission from a broadband continuum.

In conclusion, we have presented an experimental four-wave-mixing study with 16-fs pulses, which has allowed us to unambiguously extract information about the decay of the coherent emission from continuum transitions in bulk semiconductors for densities below $10^{17}\,\mathrm{cm}^{-3}$, i.e., in a density regime not accessed before. Our results demonstrate a weak density dependence and negligible energy dependence of the continuum dephasing in this density regime. We have discussed that both Coulomb and LO-phonon scattering need to be considered to describe the dephasing of continuum transitions in bulk semiconductors. A simple relaxation rate approach approximately models the experimental results. However, on the 10-fs time scale a quantum kinetics model seems more appropriate. We hope that these experimental results will further the development of a quantum kinetics model, which includes both Coulomb and LO-phonon scattering. Such a model would clarify to what extent quantum kinetics can improve the description of continuum dephasing compared to the simple relaxation-rate approach.

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