Chirp-controlled ultrafast optical nonlinearities in semiconductors

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We experimentally demonstrate that the differential transmission (DT) response of bulk semiconductors excited well above the band edge can be manipulated by chirping of the broadband excitation and readout pulses. In particular, the maximum transmission change in spectrally integrated DT experiments can be modified on the 20 fs time scale. Spectrally resolved DT studies explain this chirp dependence. Depending on the sign of the chirp, positive or negative DT contributions at low or high photon energies are probed with varying efficiency around zero time delay. These results demonstrate that chirp can become an additional degree of freedom for the optimization of device performance in ultrafast all-optical switching. © *1998 American Institute of Physics.* [S0003-6951(98)00147-8]

Time-resolved differential transmission (DT) studies have yielded a wealth of information about the dynamics of nonequilibrium carrier distributions and optical nonlinearities in semiconductors.[1] For applications in ultrafast all-optical switching, specially designed materials or material structures allow one to customize the nonlinear optical response to some extent. For example, low temperature growth reduces the carrier trapping time in GaAs[2] or electronic confinement enhances the excitonic nonlinearity in quantum wells.[3] In principle, the properties of the optical pulse can also influence light–semiconductor interaction which is referred to as coherent control.[4] In this letter, we demonstrate that the differential transmission of a bulk semiconductor can be manipulated on an ultrafast time scale by chirping of the pump and probe pulses for broadband continuum excitation. Different rises, decays, and magnitudes of the DT are observed, depending on the pulse chirp. This, in turn, leads to strikingly different switching windows. This is of particular interest for femtosecond laser applications where the broadband nonlinear optical response of semiconductor saturable absorbers creates ultrafast switching windows for pulse formation.[5] In general, switching schemes in high-speed all-optical communication systems could benefit from the controlled usage of pulse chirping, particularly if absorptive nonlinearities in semiconductors are utilized.[6] The chirp dependence of the DT response is also important in fundamental studies of carrier relaxation where a residual chirp can mask the ''true'' dynamics. Such artifacts have been observed earlier,[7,8] however, in experimental situations distinctly different from the one treated in this letter. The complete elimination of chirp becomes more and more difficult for today's sub-10 fs pulses, increasing the importance of the study of chirp effects.

Standard noncollinear pump-probe measurements were carried out with 15 fs pulses centered at 1.57 eV from a Ti:sapphire laser. Pump and probe pulses have the same spectrum, the same chirp, and are linearly parallel polarized. We have studied a 200-nm-thick $Al_{0.06}Ga_{0.94}As$ bulk semi-conductor sample with a broadband antireflection coating at the sample–air interface. The pulse spectrum spans from the band edge to states high up in the band [see inset of Fig. 1(a)]. The excitation carrier density is $N_{exc}=3\times10^{17}$ cm^{-3}. All experiments were performed at room temperature.

We can carefully control the total group delay dispersion (GDD) of our setup with double-chirped mirrors[9] and a pair of fused silica prisms. Varying the prism insertion and, therefore, the GDD, we either obtain unchirped or chirped pulses at the position of the sample. The experiments were carried out with down- or up-chirped pulses with equal pulse widths of 60 fs or with 15 fs unchirped pulses. As usual, down chirp refers to the high-energy components being in the leading
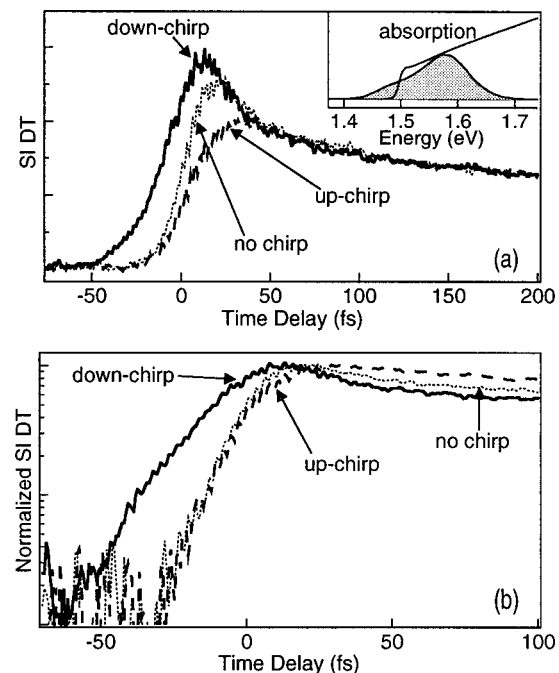

FIG. 1. Spectrally integrated differential transmission for three different pulse chirps. In the linear plot (a), the magnitude of the signals is to scale. In the logarithmic plot (b), the curves have been normalized. Inset of (a): room temperature linear absorption spectrum of the $Al_{0.06}Ga_{0.94}As$ bulk semiconductor sample and excitation pulse spectrum (shaded).
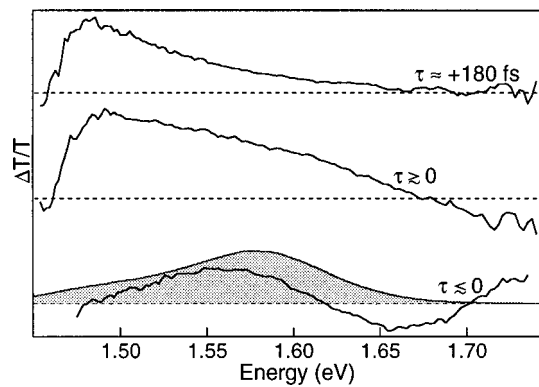
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FIG. 2. Differential transmission spectra for unchirped pump and probe pulses at three pump-probe time delays τ. Shaded: Excitation pulse spectrum.

FIG. 3. Differential transmission spectra at small negative time delay τ for up- (''red first'') and down-chirped (''blue first'') pulses. Shaded: Excitation pulse spectrum. Insets: Schematic picture of the pulse energy components vs time for up and down chirp.

edge of the pulse. The pulse widths were determined by interferometric autocorrelation and noncollinear intensity autocorrelation. They are in good agreement with the pulse widths obtained from Fourier transformation of the pulse spectrum, assuming a flat spectral phase for the unchirped pulses or a phase corresponding to the inserted GDD for the chirped pulses.

Figure 1(a) shows the spectrally integrated (SI) DT for unchirped, down-, and up-chirped pulses. For down chirp, a pronounced ultrafast decaying contribution is observed, which is weaker for unchirped and absent for up-chirped pulses. These data demonstrate that the magnitude of the nonlinear transmission can be adjusted by the pulse chirp on the 20 fs time scale. We have verified that this effect monotonically scales with the magnitude of the chirp and that it is observed over a wide range of carrier densities. For pump-probe time delays larger than 50 fs, all curves have the same height. Moreover, the rising edge of the SI DT does not reflect the pulse width, unlike the common perception. The curves for unchirped and up-chirped pulses in Fig. 1(b) show a nearly identical rise, even though the pulse widths differ by a factor of four. For down- and up-chirped pulses, the rises are different, although the pulse widths are equal. The SI DT for down-chirped pulses smears out towards negative time delay while, for up chirp, a delayed rise is observed.

To get physical insight into these chirp dependent effects, we measured the spectrally resolved (SR) DT at several delays for the three chirp conditions. We first recall the properties of the SR DT signal for unchirped pulses in Fig. 2 in order to pinpoint the chirp-dependent effects. At early time delays, we observe bleaching (positive DT) which is slightly red shifted from the excitation pulse spectrum and induced absorption (negative DT) at high energies. At later time delays, the induced absorption vanishes while the bleaching signal broadens. The center of gravity of the bleaching contributions shifts towards the band edge with time due to thermalization. Those effects are well known.[10–13] The initial red shift of the bleaching with respect to the excitation pulse spectrum and the induced absorption are consequences of many-body effects. For 100 fs pulses, they have been explained in terms of Fermi edge effects.[11] For substantially shorter pulses, the initially excited carrier distribution becomes so broad that one would not expect to see Fermi edge effects. In this case, the induced absorption
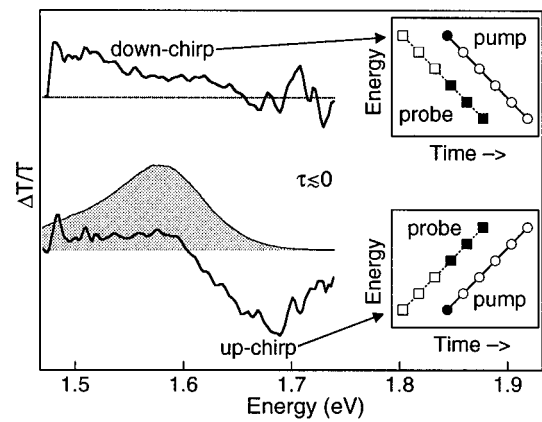
and the red shift of the bleaching were modeled by coherent nonequilibrium Coulomb enhancement arising from the local field in the coherent regime.[13]

Figure 3 shows DT spectra for up and down chirp at small negative time delay corresponding to the onset of the rise of the SI DT. The amount of positive and negative DT varies significantly compared to the unchirped situation (see Fig. 2). For down chirp, only broadband bleaching is observed while little bleaching and strongly enhanced induced absorption is obtained for up chirp. To explain these observations, we recall that efficient probing of the transmission change induced by a pump energy component is only possible with time-delayed probe components. This approach considers only the direct probe transmission term. We will comment on the grating coupling term[14] later. The perturbed free induction decay term is negligible for excitation of an inhomogeneously broadened continuum.[15]

The insets in Fig. 3 schematically show the pump and probe energy components as a function of time for both chirp directions at negative time delay. Transmission changes induced by the marked pump energy component can only be probed by the marked time-delayed probe components. Therefore, for down-chirped pulses and negative time delay, probing occurs on the low-energy side of a pump energy component. Here, only bleaching can be probed, as experimentally observed. As a consequence, the rise of the SI DT smears out towards negative time delay due to this early probing of bleaching for down chirp. For up-chirped pulses, this situation is reversed. Probing occurs on the high-energy side of a pump energy component and the induced absorption is enhanced, as seen in the experimental data. As a consequence, the rise of the SI DT is delayed because a positive integral cannot build up at early time delays.

Summarizing this discussion, the chirp results in a time delay between a pump and a probe energy component which depends on their energy separation. Since this time delay determines the probing efficiency, an energy-dependent readout efficiency results.

The energy dependence of the readout efficiency also explains the DT data at later times. Figure 4 shows DT spectra for down and up chirp for a small positive time delay corresponding to the maximum of the SI DT. As the insets
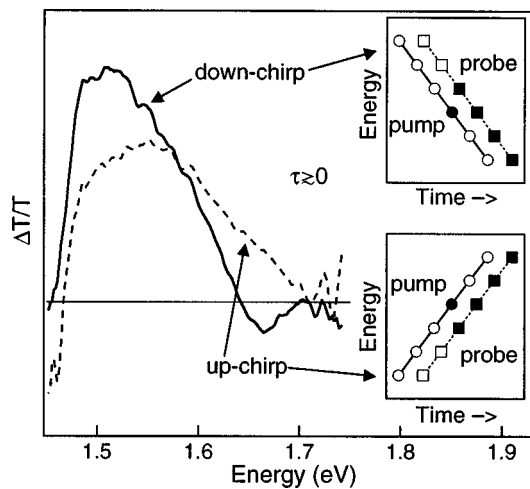
FIG. 4. Differential transmission spectra at small positive time delay τ for up- (red first) and down-chirped (blue first) pulses. Insets: Schematic picture of the pulse energy components vs time for up and down chirp.

illustrate, for up chirp, probing at high energies is still most efficient. Therefore, enhanced DT is observed at high energies. For down chirp, probing at low energies is most efficient, leading to enhanced DT close to the band edge. Because the bleaching signal shifts with time towards the band edge, enhanced probing at low energies results in a higher spectrally integrated differential transmission than enhanced probing at high energies. This manifests itself experimentally in the pronounced fast contribution to the SI signal for down-chirped pulses, as shown in Fig. 1.

The above discussion has shown that the observed chirp dependence of the SI DT signal can be traced back to the shape of the DT spectra. This shape can be explained by a qualitative model which considers the chirp-induced energy dependence of the readout efficiency. Intrinsically, these considerations only take into account changes in the direct probe transmission. However, during pulse overlap, the grating coupling term[14] also contributes to the DT. This has been neglected so far. The grating coupling term arises from the diffraction of the pump from the grating formed by the pump and the probe. We have experimentally verified that this term does not dominate the observed chirp effects. For this purpose, we have weakened the grating coupling term by changing the pump-probe configuration from linear parallel polarizations to linear perpendicular polarizations.[16] We still observe the same influence of the chirp on the DT. Consequently, we conclude that the dominant effects due to chirp result from the probe transmission term, in agreement with our qualitative model.

These results do not contradict the work reported in Ref. 7, where it has been shown that the grating coupling term strongly depends on the chirp in a configuration where only the pump pulse is chirped. We have experimentally verified that the DT response significantly changes if both the pump and the probe pulse are chirped. Therefore, it does not come as a surprise that different terms are important for the chirp effects in the different experimental situations. In fact, the additional probe chirp is the reason for the importance of the direct probe transmission term because this probe chirp leads to the chirp-induced energy-dependent readout efficiency.

In summary, we have shown that the differential transmission of a bulk semiconductor can be manipulated by chirping of 15 fs pulses. Our data demonstrate that the transmission can be increased in 20 fs-wide time windows by appropriate chirping, allowing for ultrafast switching with increased modulation. Spectrally resolved studies show that this is due to the chirp-induced selective readout of bleaching and induced absorption during carrier thermalization.

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