

Laser-Induced Above-Band-Gap Transparency in GaAs

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We report the observation of large ($\sim 40\%$) laser-induced above-band-gap transparency in GaAs at room temperature. The induced transparency is present only during the pulse width of the driving midinfrared laser pulses and its spectral shape is consistent with a laser-induced blueshift of the band edge. Our simulations based on the dynamic Franz-Keldysh effect reproduce the salient features of the experimental results, demonstrating, in particular, that the amount of the band edge shift is approximately given by the ponderomotive potential.

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Under certain conditions, a normally opaque medium can be made transparent through coherent interaction with laser light. Such laser-induced transparency phenomena include self-induced transparency [1] and electromagnetically induced transparency [2], which have been extensively studied in atomic and molecular systems. Solids, semiconductors, in particular, are an obvious alternative to gases for further exploring such coherent light-matter interactions. The direct interplay of the spatially periodic lattice potential and the temporally periodic laser field is predicted to cause drastic band structure modifications such as band gap distortion and oscillations [3,4], dynamic localization [5], and the appearance of new gaps [6]. In addition, semiconductors exhibit various many-body effects [7] and hence become an even richer system for such studies. Furthermore, the effect of quantum confinement is predicted to exhibit exotic quantum dynamics, including coherent destruction of tunneling [8], collapse of minibands [9], and quantum chaos [10]. However, these phenomena have been mostly unexplored experimentally due to the high required laser intensities and unavoidable sample damage.

Here, we demonstrate a new type of laser-induced transparency effect in semiconductors. The key to this effect is the use of intense *midinfrared* (MIR) light, which helps minimize interband absorption and sample damage while maximizing the ponderomotive potential U_p (or the “wiggle” energy) [11,12], which increases quadratically with increasing wavelength. For a driving laser field with vector potential $A = A_0 e^{i\omega t}$,

$$U_p = \frac{e^2 A_0^2}{4mc^2} = \left(\frac{2\pi e^2}{mc} \right) \left(\frac{I}{\omega^2} \right), \quad (1)$$

where I is the intensity of the laser field, e is the electronic charge, c is the speed of light in vacuum, and m is the electron mass. When U_p becomes comparable to or larger than a characteristic energy of the system (such as the ionization potential), *extreme* nonlinear optical effects are expected [13–15]. Under such conditions we observed

ultrafast photo-induced transparency right above the band edge of GaAs at room temperature. The effect can be interpreted as a laser-induced blueshift of the band edge, whose amount is given by U_p . In addition, photo-induced absorption below the band edge with unusual laser power dependence was observed. We explain these observations in terms of the dynamic Franz-Keldysh effect (DFKE) [16–18].

The DFKE is an intense-field effect in which changes are induced in the transmission spectra near the fundamental band edge of a semiconductor in the presence of an intense, high frequency field [16–18]. These changes include absorption below the band edge, a blueshift of the band edge leading to induced transparency, and oscillatory behavior above the band edge (see Fig. 1). Also, nonlinear mixing between the strong pump and a weak probe beam leads to the generation of optical sidebands [15,18–21]. The DFKE, which is a nonresonant (far from resonance) effect, is quite different from the ac Stark effect which occurs in the presence of strong driving fields resonant (or nearly resonant) with electronic transitions. Hence, the DFKE is an ultrafast virtual process in which no real carriers are created in the sample. Such nonresonant, nonperturbative phenomena are observed when the applied field is at the transition between the classical and quantum regimes, i.e., when the interaction with the applied field cannot be treated by either neglecting photon effects or by considering only single photon effects [12]. Recently, Nordstrom *et al.* reported an *excitonic* DFKE in quantum wells using a terahertz free electron laser [18] while Chin *et al.* observed below-band-gap absorption and sideband generation in bulk (optically-thick) GaAs using intense picosecond MIR pulses [12,15]. However, neither the predicted blueshift of the absorption edge nor the oscillatory behavior in the spectrum *above the band edge* has been clearly observed in the previous studies. In order to minimize the excitonic effects which could prevent us from clearly observing the DFKE, we chose to study bulk GaAs samples at room temperature.

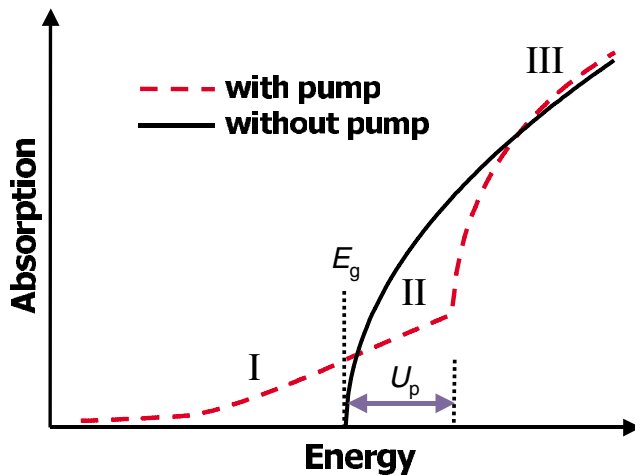


FIG. 1 (color online). Interband absorption in a typical direct-gap semiconductor near the band gap E_g with (dashed line) and without (solid line) a strong driving field. The dynamic Franz-Keldysh effect predicts: (i) below-band-gap absorption (region I), (ii) blueshift of the band edge causing an induced transparency (region II), and (iii) oscillatory behavior above the band gap (region III). The magnitude of the blueshift is predicted to be equal to the ponderomotive potential U_p .

We performed a pump-probe study of the transmission near the band edge of bulk GaAs samples using an optical parametric amplifier system with difference-frequency mixing as a source of tunable MIR pump pulses. The system produced MIR pulses (~ 150 fs) tunable from 3 to 20 μm at a 1 kHz repetition rate and was pumped by a Ti:sapphire-based chirped pulsed amplifier (Clark CPA-2010). A broadband continuum in the form of white light was used as the probe beam. The white light was produced by continuum generation in a sapphire plate, using a small fraction of the CPA beam. The broadband probe and the MIR pump beams were then focused onto the sample using an off-axis parabolic mirror. A computer-controlled delay stage was used to temporally overlap the two pulses. After passing through the sample, the spectrum of the probe beam was dispersed using a grating monochromator and detected using a Si charge-coupled device camera.

We first tried to observe the below-band-gap absorption predicted by the theory. For this, a semi-insulating bulk GaAs (band gap ~ 1.42 eV) wafer with a thickness of ~ 350 μm served as sample. Shown in Fig. 2 is the normalized transmission of the probe beam in the presence of the pump beam below the band edge as a function of photon energy. As can be seen from the graph, there is a huge absorption of the probe beam below the band edge which leads to almost complete quenching of the transmission (transmission without the pump on the same scale is about 0.5). This decrease in transmission due to induced absorption occurs only during the temporal overlap of the pump and the probe pulses, confirming the

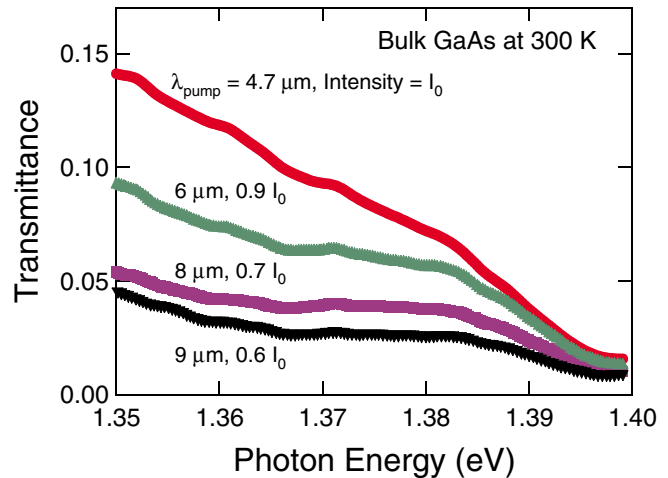


FIG. 2 (color online). Transmission quenching below the band gap in the presence of strong MIR pump beams of different wavelengths. The effect increases with increasing wavelength despite the fact that the intensity of our MIR source decreases for longer wavelengths. This is because the ponderomotive potential is larger for longer wavelengths in our case.

effect to be a virtual one, i.e., no real carriers and/or lattice-heating effects are involved. Furthermore, strikingly, the quenching is more pronounced for longer wavelengths in spite of the fact that the optical parametric amplifier output intensity decreased for higher wavelengths. This counterintuitive observation can be explained only by considering the fact that the ponderomotive potential increases for higher wavelengths due to the $1/\omega^2$ dependence [see Eq. (1)].

Any multiphoton interband absorption of the pump photons is ruled out here as this would require the Keldysh parameter γ [22], defined as

$$\gamma = \frac{\omega(mE_g)^{1/2}}{eE}, \quad (2)$$

to be much larger than unity. Here, E_g is the band gap and E is the electric field. In our case, however, we estimate γ to be about 1.8 and hence we are far away from the multiphoton regime. Also, multiphoton interband absorption would imply generation of real carriers whose effect would remain even after the pump and probe pulses do not overlap in time. Thus, we conclude that the observed below-band-gap absorption is due to the photon-assisted tunneling of electrons to the conduction band and can only be attributed to the DFKE.

In order to observe laser-induced modifications in transmission spectra above the band edge, a GaAs film sample (~ 2.3 μm thick) was used. Figure 3(a) shows typical data showing induced transparency. Here the black trace represents the transmission of the white light probe beam through the film sample in the presence of 9 μm MIR pump of peak intensity $\sim 10^9$ W/cm^2

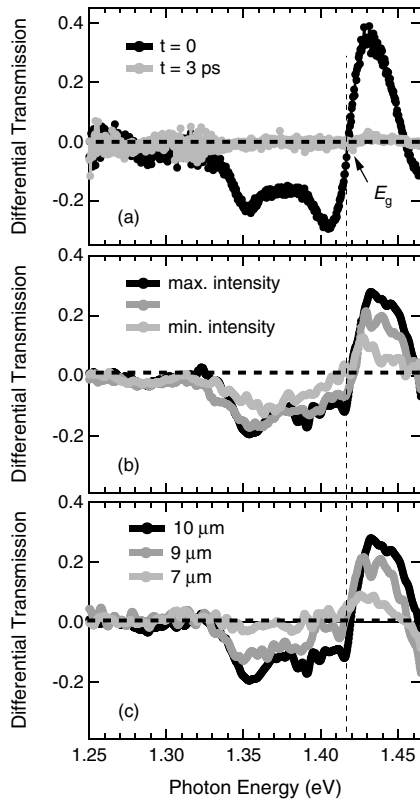


FIG. 3. (a) Normalized transmission spectra of the white light probe beam through a GaAs film for zero time delay (black) and a 3 ps time delay (gray). The pump wavelength was $9 \mu\text{m}$. A large ($\sim 40\%$) induced transparency is observed above the band gap (1.42 eV) only when the two beams overlap temporally. (b) The observed change in transmission spectra of the probe beam in the presence of the pump beam ($9 \mu\text{m}$) with different intensities. (c) Transmission spectra of the probe beam for different pump wavelengths. The intensity of the pump beam decreases with increasing wavelength. The observed effect is more pronounced at longer wavelengths, even though the intensity of the pump beam decreases with increasing wavelength.

($U_p/\hbar\omega \sim 0.13$) when the pump and probe coincide temporally; data are normalized to the transmission in the absence of the pump. A large ($\sim 40\%$) induced transparency is observed right above the band edge (1.42 eV). Also shown in Fig. 3(a) is a gray trace, which was taken when the pump arrived at the sample 3 ps before the probe and shows no induced transmission changes. A detailed time-dependence study (not shown) indicated that the photo-induced changes (including the transparency) exist only when the pump and probe pulses overlap temporally. This fact demonstrates, once again, the virtual nature of the effect.

In Fig. 3(b), intensity dependence for a fixed pump wavelength ($9 \mu\text{m}$) is shown. As expected, the photo-induced transparency decreases for lower intensities of

the pump beam. Finally, the dependence of the effect on the pump wavelength was studied [Fig. 3(c)]. The effect is better resolved at longer wavelengths, a trend similar to the below-band-gap case. We did not observe a significant change in the peak position of the induced transparency for different wavelengths as we expect $U_p/\hbar\omega$ to be similar in magnitude (~ 0.1) for all the cases. However, a careful examination of Figs. 3(b) and 3(c) shows slight shifts of the peaks when intensity and wavelength of the pump beam are varied.

The existing models predict an exponential tail (apart from the Urbach tail) in the absorption spectra below the band gap and oscillations above the band gap. Also, a blueshift of the absorption edge by an amount equal to U_p is predicted. In order to compare our data with the theory, we use Yacoby's model [16]. It calculates the electronic energy levels of a bulk semiconductor in the presence of a strong pump beam of frequency ω , which is treated non-perturbatively. The transition rate between two such states due to an additional perturbation of frequency Ω (probe beam) is then found. In our experiments, we measured the time-averaged transmission of the probe beam and hence we calculated the time-averaged transition rates for valence band to conduction band transitions using Yacoby's theory [see Eq. (38) in [16]]. Any excitonic contributions were assumed to be negligible as we are dealing with a bulk semiconductor at room temperature. The values of intensity, wavelength, and effective mass for the calculation were taken to be $I = 10^9 \text{ W/cm}^2$, $\lambda = 9 \mu\text{m}$, and $m^* = 0.067m_e$, respectively, corresponding to $U_p = 0.13\hbar\omega$. From the time-averaged change in transition rate, change in the real and imaginary parts of the dielectric constants were calculated using Kramers-Kronig relation. Finally, as our sample was thin enough to cause interference effects by multiple reflections, we calculated the change in transmission by explicitly taking such effects into account (see, e.g., [23]). Such Fabry-Perot type interference is important mainly below the band gap where absorption is fairly small.

The calculated and measured differential transmission spectra are plotted in Fig. 4. For comparison with the theory, the abscissa of the plot was changed from $\hbar\Omega$ to $\hbar\Omega - E_g$, where E_g is the band gap of GaAs and expressed in units of $\hbar\omega$. There is good agreement with the theory given the fact that we did not use any fitting parameters. Most importantly, we observe that the large photo-induced transparency is in fact due the blueshift of the absorption edge. The magnitude of the blueshift is equal to our estimate of U_p ($0.13\hbar\omega$) as is expected from the theory. However, it should be mentioned here that we have scaled down the above-band-gap peak by a factor of 6. This is justified because in our calculation we have not included any Urbach-type tail arising from phonons and impurities in the sample [24,25]. Such a tail would actually broaden and decrease the peak above the band gap.

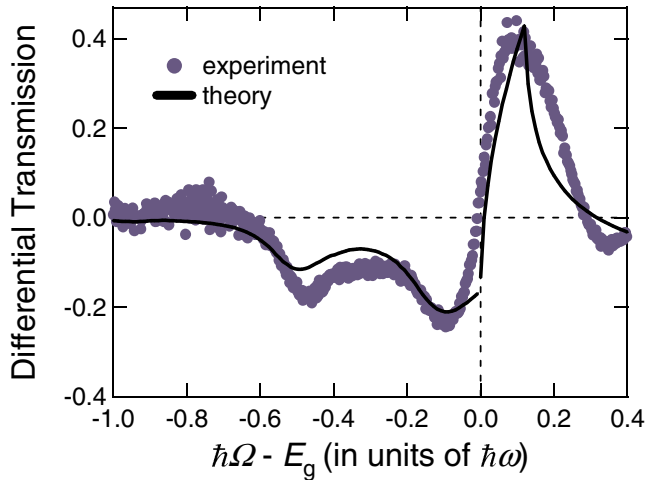


FIG. 4 (color online). Theoretical fit to the experimental data based on Yacoby's theory (see text).

In conclusion, we have made the first observation of large ultrafast induced transparency above the band edge and also huge absorption below the band edge of GaAs due to the dynamic Franz-Keldysh effect. We understand this novel transparency as a result of the blueshift of the band edge. The existing theory is able to explain our experimental observation reasonably well and yields an accurate value for the magnitude of the observed blueshift.

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