Femtosecond Photon Echoes from Band-to-Band Transitions in GaAs

P. C. Becker, H. L. Fragnito,[a] C. H. Brito Cruz,[a] R. L. Fork, J. E. Cunningham, J. E. Henry,
and C. V. Shank
AT&T Bell Laboratories, Holmdel, New Jersey 07733

(Received 19 May 1988)

We report the first observation of femtosecond photon echoes from the band-to-band transitions in a bulk semiconductor. The time decay of the echo, found to vary from 3.5 to 11 fs, has allowed us to determine the polarization dephasing rate in GaAs. This rate was found to depend on the carrier density in the experimental range covered, 1.5×10^{17} to 7×10^{18} cm^{-3}, indicating the dominance of carrier-carrier scattering as the principal dephasing mechanism. The observed functional dependence of the dephasing rate on the carrier density has yielded previously unavailable information on Coulomb screening in a nonequilibrium carrier distribution.

PACS numbers: 72.20.Jv, 78.47.+p

The photon-echo[1] or time-delayed four-wave mixing technique has become an important tool for investigating dephasing processes in gases,[2] solids,[3,4] and glasses.[5] The use of coherent optical transients to study such processes for band-to-band transitions in semiconductors has been frustrated by the rapid time scale on which such processes occur. Recent advances in short pulse techniques which have led to the generation of optical pulses as short as 6 fs[6] have made such investigations possible. With this increased time resolution we have been able to make the first observation of two-pulse photon echoes from direct transitions in GaAs and have determined the polarization dephasing rate.

The polarization dephasing rate measured in the experiments described here provides a direct measure of the process of momentum dephasing.[7] At high carrier densities the carrier momentum loses phase coherence primarily through the screened Coulomb interaction between carriers. Both elastic and inelastic carrier-carrier collisions contribute to the momentum dephasing. At low carrier densities electron-phonon interactions begin to dominate in the intrinsic material. The density dependence of the polarization dephasing rate provides important information concerning the carrier-carrier interaction.

In the experiments reported here we observe photon echoes using a two-pulse sequence. Two pulses, one having wave vector $\mathbf{k}_1$ and the other wave vector $\mathbf{k}_2$, generate an echo in the momentum-matched direction $2\mathbf{k}_2-\mathbf{k}_1$. The angle between $\mathbf{k}_1$ and $\mathbf{k}_2$ is small. The echo is then separated spatially from the exciting pulses. Since there is a sizable frequency spread in each pulse due to its short duration, there is an angular spread of the $\mathbf{k}$ vectors that make up the echo signal. However, the entire echo signal, which is well separated spatially from the incident beams, is collected by our detection optics. The energy of the generated echo is measured as a function of the relative time delay between the exciting pulses. The sample is at room temperature.

We can model the band-to-band absorption in a direct semiconductor such as GaAs as a set of Lorentzian two-level transitions having a half-width of $2/T_2$. We can write the band-to-band absorption coefficient $\alpha(E)$ as

$$\alpha(E)=\int dE'\frac{\rho(E')}{(E-E')^2+(2\hbar/T_2)^2}, \tag{1}$$

where E is the absorption energy, $\rho(E')$ is the density of states at energy E', $\hbar$ is Planck's constant divided by 2π, and T_2 is the polarization dephasing time. With the above model for the absorption, the energy of the echo can be determined to vary exponentially with the relative time delay τ between the two pulses as

$$E(\tau)\propto\exp(-\tau/T_{\text{echo}}), \tag{2}$$

where $T_{\text{echo}}=T_2/4$.[8] Thus by measuring the echo energy as a function of time the polarization dephasing time T_2 can be directly determined.

The experiment was performed with use of compressed pulses phase corrected to third order in a manner described previously.[6] The duration of the excitation pulses was measured to be in the range 6 to 10 fs with use of the second-harmonic up-conversion technique. The pulse repetition rate was 8 kHz and the pulse energy was on the order of 1 nJ. The energy of the pulse was much less than that needed for a π pulse so the echoes observed in the experiments described here are in the small-signal perturbation limit. The pulses were split with use of a modified Michelson interferometer configuration to form the two excitation pulses. The two pulses were focused with a 5-cm focal length lens into a 0.1-μm thick sample of GaAs grown by molecular-beam epitaxy. Both faces of the sample were antireflection coated. The excitation pulse energy at the sample ranged from 0.1 to 0.01 nJ per pulse which corresponds to carrier densities ranging from 10^{17} to 10^{18} cm^{-3}. The carrier density was estimated by measuring the number of photons absorbed in the material. The spot size of the focused beam was measured to be 30 μm in diameter.

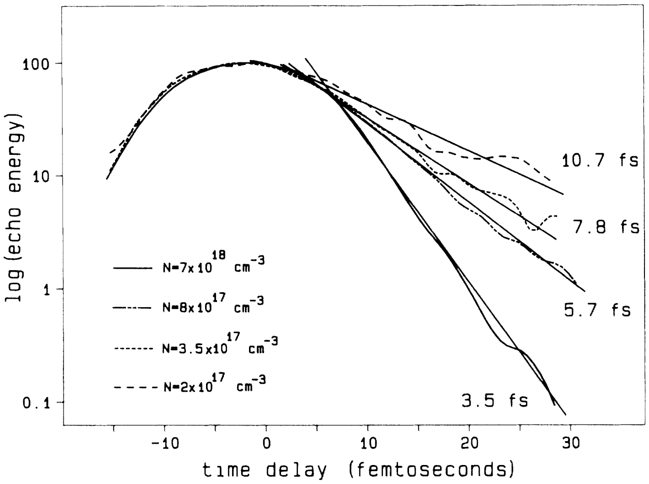

FIG. 1. The photon-echo signal in GaAs at room temperature is plotted as a function of relative time delay between the two 6-fs exciting pulses. The time constant of the exponential decay, T_{echo}, is indicated for each carrier density.

The signal was detected in the $2\mathbf{k}_2 - \mathbf{k}_1$ direction with a lens spatial filter to reject stray light. The selected signal was then directed into a photomultiplier. The detection electronics consisted of a boxcar integrator followed by a phase-lock detector. The signal was recorded as a function of relative time delay τ between the pulses.

In Fig. 1 we have plotted the log of the echo energy versus the relative time delay between the two pulses. At the highest density an exponential decay with a time constant of 3.5 fs is measured. This is very close to the system response limit. As the density is reduced the echo decay time lengthens and is very clearly resolved.

The echo decay constant, T_{echo}, is plotted as a function of density in Fig. 2. The points are experimental and the solid curve is a power-law fit to the data and is given by the expression

$$T_{\text{echo}} = 6.8 N^{-0.3}. \qquad (3)$$

According to expression (1) the polarization dephasing time is given by the relation $T_2 = 4T_{\text{echo}}$. The data reveal that we observe the dephasing time to range from 14 to 44 fs while the density has been changed from 7×10^{18} to 1.5×10^{17} cm^{-3}. These times are significantly shorter than those measured by Oudar *et al.* in their experiment on the relaxation of induced anisotropy in GaAs for near band-edge carriers. The dependence of the dephasing rate on density clearly suggests that the dephasing process is dominated by carrier-carrier interactions. Carrier-phonon scattering has also been shown to be density dependent through screening of the Frohlich interaction. However, the experimental data presented here show a rate that increases with density whereas the screened electron-phonon scattering rate decreases with density.[9]

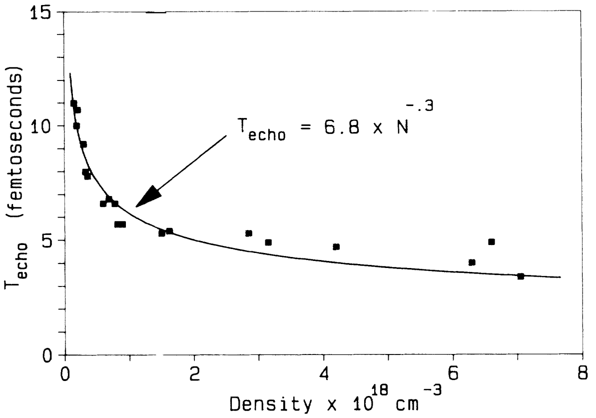

FIG. 2. Echo decay time constant as a function of the carrier density, for GaAs at room temperature.

The momentum relaxation rate T_m in the limit of large carrier densities is given by

$$T_m \propto N^{-1} S(N), \qquad (4)$$

where N is the carrier density and $S(N)$ is a density-dependent factor representing the effect of Coulomb screening. In the Thomas-Fermi approximation $S(N) \approx N^{2/3}$, which is close to the experimentally measured $S(N) \approx N^{0.7}$. However, the Thomas-Fermi approximation is not valid in this situation because the excited carrier distribution is nonthermal. Currently a good theory does not exist to describe the process of screening in this highly nonequilibrium population distribution. These experiments provide the first information on nonequilibrium screening processes on such a rapid time scale.

The echo relaxation time is seen to increase as the excitation density is lowered. As the dephasing time becomes comparable to the electron-phonon interaction time (on the order of 100 fs), phonon processes are expected to begin to dominate and T_{echo} should become independent of excitation density. In these experiments we have not reached that limit because the echo signal becomes too weak to detect.

The polarization of an electron-hole pair can be dephased by collisions involving both electrons and holes. The scattering processes for both types of carriers have to be taken into account in any calculation or numerical simulation of carrier dephasing under the experimental conditions described here. Levi and co-workers have calculated scattering rates for a single hot electron injected into a sea of carriers in the $T = 0$ limit, including the effects of screening and of scattering from coupled plasmon-phonon modes.[10,11] Their calculated scattering rates for electron-electron and electron-hole scattering are comparable to the rates measured in the present photon-echo experiment, even though the experimental carrier energy distribution is quite different from that assumed in the calculations.

In conclusion, we have measured the decay of photon echoes in GaAs from band-to-band transitions. These measurements open the way to a new class of coherent transient investigations in semiconductors. The density dependence of the dephasing rate challenges our understanding of Coulomb screening of nonequilibrium carrier distributions on an unprecedented time scale.

We are grateful to S. Schmitt-Rink, D. S. Chemla, and J. Shah for valuable discussions, and to D. W. Taylor and F. A. Beisser for expert technical contributions.

---

[a] On leave from Instituto de Fisica, University of Campinas, Campinas 13100 SP, Brazil.

[1] N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Lett. **13**, 567 (1964).

[2] W. H. Hesselink and D. A. Wiersma, Phys. Rev. Lett. **43**, 1991 (1979); R. M. Macfarlane, R. M. Shelby, and R. L. Shoemaker, Phys. Rev. Lett. **43**, 1726 (1979).

[3] C. K. N. Patel and R. E. Slusher, Phys. Rev. Lett. **20**, 1087 (1968); J. P. Gordon, C. H. Wang, C. K. N. Patel, R. E. Slusher, and W. J. Tomlinson, Phys. Rev. **179**, 294 (1969).

[4] L. Schultheis, J. Kuhl, A. Honold, and C. W. Tu, Phys. Rev. Lett. **57**, 1635 (1986).

[5] J. Hegarty, M. M. Broer, B. Golding, J. R. Simpson, and J. B. MacChesney, Phys. Rev. Lett. **51**, 2033 (1983).

[6] R. L. Fork, C. H. Brito Cruz, P. C. Becker, and C. V. Shank, Opt. Lett. **12**, 483 (1986).

[7] J. L. Oudar, A. Migus, D. Hulin, G. Grillon, J. Etchepare, and A. Antonetti, Phys. Rev. Lett. **53**, 384 (1984).

[8] T. Yajima and Y. Taira, J. Phys. Soc. Jpn. **47**, 1620 (1979).

[9] S. Das Sarma, J. K. Jain, and R. Jalabert, Phys. Rev. B **37**, 6290 (1988).

[10] A. F. J. Levi, J. R. Hayes, and R. Bhat, Appl. Phys. Lett. **48**, 1609 (1986).

[11] A. F. J. Levi and Y. Yafet, Appl. Phys. Lett. **51**, 42 (1987).