Two-Dimensional Carrier-Carrier Screening in a Quantum Well

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The behavior of carrier-carrier screening is investigated in a GaAs-GaAlAs quantum-well structure by measuring the band-to-band polarization dephasing with femtosecond photon echoes. The variation of the electron-hole-polarization dephasing time with the carrier concentration reveals the two-dimensional character of the short-range screening between the interacting carriers.

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The phase coherence time between an electron and the corresponding hole created during a band-to-band optical transition in a semiconductor decreases with increasing carrier density due to the effects of carrier-carrier scattering. At low carrier densities, ignoring the Coulomb interaction between carriers, the carrier-carrier scattering time is expected to scale inversely with the carrier density. At higher densities, screening due to the Coulomb potential of the charged particles will act to suppress the process of carrier-carrier scattering. Coulomb screening, which was originally studied in metals [1], depends on the electron and hole environment and therefore is a function of the dimensionality of the system under study.

We report here measurements that show the influence of carrier-carrier interactions on the dephasing processes which occur in a GaAs-GaAlAs quantum-well (QW) structure. Because of the confinement of the carriers in the well, they are expected to behave like a two-dimensional (2D) electron gas. Indeed, our measurements performed at different carrier densities indicate a screening behavior that reveals the bidimensionality of the system. The 2D screening that we deduce from the experiment is well understood if we assume that a given carrier interacts only with its nearest neighbors, indicating that the range of the screened Coulomb interaction is of the order of the average intercarrier spacing. We discuss the validity of this nearest-neighbor interaction picture with respect to the experimental conditions in which a nonequilibrium, nondegenerate electron-hole population is created over a large range of energies and wave vectors.

Femtosecond optical spectroscopy now allows the dynamics of energy relaxation and dephasing of excited carriers in bulk semiconductors or 2D QW structures to be studied with 10-fs resolution [2]. In bulk GaAs it is known that under high densities of excitation, carrier-carrier scattering plays an important role in the thermalization of hot carriers (carriers with high excess energy with respect to the bottom of the conduction or valence band [3]), and it has been shown with both photon-echo [4] and time-resolved polarization rotation measurements [5,6] that it is also the dominant mechanism for momentum redistribution. In QW structures the carrier thermalization has been extensively studied in undoped [7,8] and doped [9,10] structures, where the importance of collisions between carriers was found to be the main process for the redistribution of energy through inelastic carrier-carrier scattering.

Recent work [8-10] has also shown that the phase-space filling due to the Pauli exclusion principle dominates over the long-range Coulomb screening in bleaching of the exciton. This was clearly demonstrated by the appearance of exciton bleaching only when the carriers occupy the lowest-energy states of the first QW subband. In addition to these works, the dephasing of free excitons in bulk GaAs and GaAs-GaAlAs structures has been studied and proved to be related to exciton-phonon scattering for low excitation densities and to exciton-exciton scattering for high excitation densities [11]. Until now, there have been no direct temporal measurements of the carrier-carrier dephasing time in a 2D QW. The Coulomb screening is expected to be important for the momentum redistribution during the time scale when the carriers have not completely thermalized. The observation of this effect, which we investigate here with photon echoes, requires a temporal resolution of less than 10 fs.

The generation of photon echoes with femtosecond pulses has already proven to be a very powerful experimental technique for measuring polarization dephasing times in molecules in solution as well as in semiconductors [12]. Using this technique, dephasing times in the range 15-50 fs have been reported in bulk GaAs [4]. Moreover, the variation of the dephasing time with the three-dimensional carrier density N was shown to follow a power law, $T_{\text{echo}} \sim N^{-0.3}$, characteristic of a three-dimensional environment. In our experiment, we use this technique with 8-10-fs near-infrared pulses to study a 2D environment in a 9.6-nm-thick GaAs quantum well at room temperature.

The ultrashort infrared pulses are generated as follows [13]. The 50-fs pulses coming from a CPM dye laser (620 nm) are amplified to a few microjoules by a copper-vapor laser at an 8-kHz repetition rate, and are focused in a jet of ethylene glycol to generate a white-light continuum. The near-infrared part (at 805 nm) of this spectral continuum is selected with a slit and amplified in a gain medium (LDS-821 in ethylene glycol) with a second copper-vapor laser. The resulting 50-fs infrared pulses, which can be tuned within the LDS-821 gain response curve, are further shortened to 10 fs using a fiber and a sequence of gratings and prisms as previously described [14].

The photon echoes are produced by the compressed pulses which are split into two parts in a Michelson interferometer and focused on the QW structure to a ~30-μm spot. The pair of femtosecond pulses, with respective wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, create a photon echo in the phase-matched $(2\mathbf{k}_2-\mathbf{k}_1)$ direction [15]. The energy of the echo is detected with a photomultiplier, box-car integrator, and lock-in amplifier, for different relative delays between the two incident pulses. The resulting decay time of the echo signal T_{echo} is a direct measurement of the polarization dephasing time T_2, since $T_{\text{echo}}=T_2/4$ for an inhomogeneously broadened system (in the limit of infinite inhomogeneity).

Figure 1 shows the optical density of the QW structure used in these measurements. The spectrum (730–900 nm total bandwidth) of the 8-fs optical pulses is also shown for comparison. It can be seen that the pulse spectrum overlaps the 2D absorption region of the QW structure, including the $n=1$ and $n=2$ excitons. Under these conditions, the ratio between the density of carriers excited in the 3D continuum of the QW structure and those excited in the well is estimated to be of the order of 6%. This estimation is made by assuming that the optical density in Fig. 1 is the sum of a pure 2D carrier population and a 3D population. The 3D component is deconvolved from Fig. 1 by extrapolation of its short-wavelength spectrum (taken for $660\text{ nm}<\lambda<720\text{ nm}$) to the long-wavelength region ($720\text{ nm}<\lambda<860\text{ nm}$). The percentage of 3D carriers is then calculated using the ratio

$$\frac{\int I_i(\omega)\{1-\exp[\alpha_{3\text{D}}(\omega)l]\}d\omega}{\int I_i(\omega)\{1-\exp[\alpha_{\text{meas}}(\omega)l]\}d\omega},$$

where $I_i(\omega)$ is the incident pulse spectrum, and $\alpha_{3\text{D}}(\omega)l$ and $\alpha_{\text{meas}}(\omega)l$ are respectively the 3D optical density (known from the above deconvolution) and the measured optical density.

This estimation, deduced from the experiment, represents an average of the carrier confinement. It has the advantage of taking into account the real sample absorption, including all the processes contributing to the 3D component (impurity scattering, temperature band-gap broadening, etc.). However, it does not distinguish between the contributions of electrons, heavy holes, or light holes in the different subbands. The degree to which carriers are confined to two dimensions will of course depend on the effective mass, as well as the excess energy of the carriers relative to the band gap. The quantum wells used in this experiment are GaAs-$\text{Ga}_{0.7}\text{Al}_{0.3}\text{As}$ with 96-Å wells and 98-Å barriers. For electrons and heavy holes in the $n=1,2$ subbands, and light holes in the $n=1$ subband, the confinement in the well (probability of finding the particle in the well region) is in excess of 80%. For the $n=2$ light holes, the confinement is ~75% [16]. Thus the QW structure provides good 2D confinement of the carriers over our range of excitation, and our experimental results are consistent with what one would expect from a 2D gas with Coulomb screening.

In Fig. 2, we have represented the logarithm of photon-echo signals from the QW structure as a function of the relative delay between the two infrared pulses. T_{echo} is obtained from the exponential decay of the corresponding echo signal using a least-squares linear fit. When the carrier density is increased from 10^{10} to 10^{11} cm^{-2}, the echo decay time decreases from 50 to 16 fs indicating the effects of carrier-carrier scattering on the electron-hole dephasing process observed in these measurements. Temporal structure in the echo signal at the lowest carrier densities (dotted curve in Fig. 2) is within the noise level of the system and is not physically interpretable.

Figure 3 shows the echo decay time T_{echo} measured for different carrier densities N. For each density in this curve, T_{echo} is obtained from the exponential decay as in Fig. 2. The experimental error bar for each point indicates the good signal-to-noise ratio. The solid curve is a

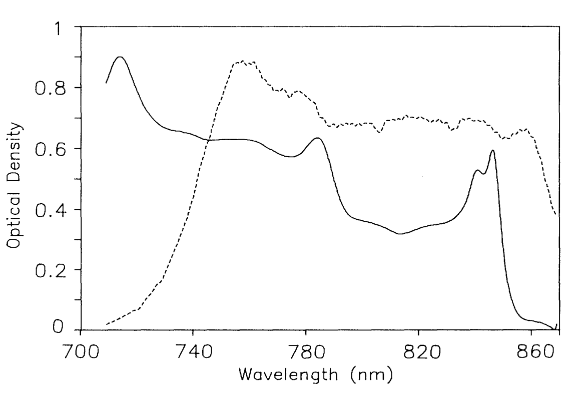

FIG. 1. Optical density of the 9.6-nm-thick QW (solid line) and spectrum of the pulse (dashed line).

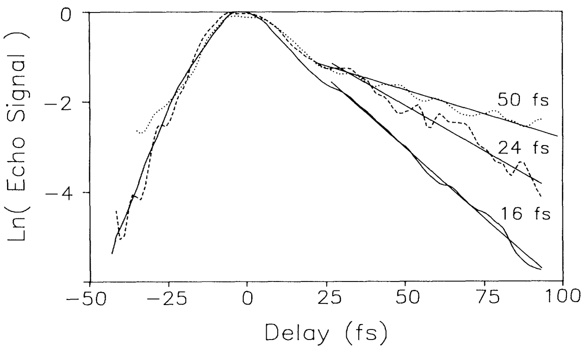

FIG. 2. Photon echo from the QW structure as a function of time delay for different carrier densities N: dotted line ($N=10^{10}$ cm^{-2}), dashed line ($N=3.4\times10^{10}$ cm^{-2}), solid line ($N=10^{11}$ cm^{-2}).

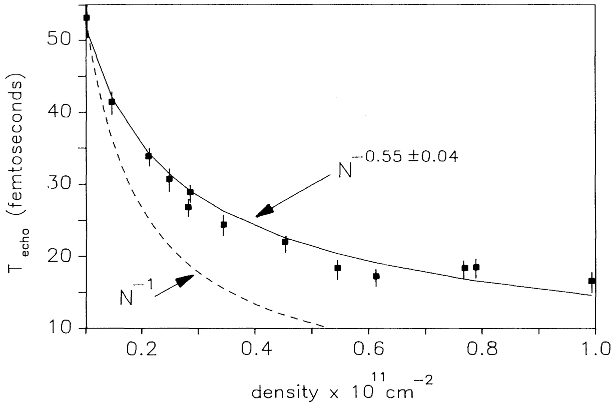

FIG. 3. Variation of the echo decay time with the carrier density. The solid line is a regression fit which gives a power law proportional to $N^{-0.55 \pm 0.04}$. The dashed line is the predicted density dependence without the effects of Coulomb screening.

fit to the data which gives the dependence of the dephasing time on carrier density with an exponent of -0.55 ± 0.04. Since the experimental technique that we use is sensitive to the phase relaxation, both elastic- and inelastic-scattering processes are involved. The fast decay times deduced from our data are consistent with the fact that the phase coherence between an electron and a hole is lost long before the carrier thermalization is reached. Thermalization of an initial nonthermal carrier distribution is known to occur on a time scale of 200 fs in similar QW structures [8].

The dashed line in Fig. 3 indicates the dependence of the electron-hole dephasing on carrier density predicted by a kinetic gas (Drude) model. If the Coulomb interaction between the carriers is not considered, then the carrier-carrier scattering time (i.e., electron-hole dephasing time) will scale as N^{-1}. Deviation from this scaling is attributed to the effects of Coulomb screening. The exponential dependence, $N^{-0.55}$, observed in our measurements reflects the two-dimensional screening behavior of the carriers. A similar experiment performed on a bulk sample of GaAs gave an exponent of -0.3 for the density law [4]. Indeed, the Coulomb interaction between two charges is less screened in a 2D particle gas than in a 3D gas, since due to the confinement of carriers in a layer, the environment is less effective in shielding the attractive or repulsive forces. The behavior observed here is consistent with a screened Coulomb interaction having a very short effective range. This is known to occur in systems with high carrier densities [17], where the range of the Coulomb interaction is much reduced due to the mutual screening of the charged particles.

We can model our experimental results, as well as those reported in [4], using very simple geometrical arguments based on short-range interactions. For a system with dimensionality d, considering nearest-neighbor interactions, the scattering rate of a charge in a "sphere" containing N other charges scales as $N^{1/d}$. Therefore, the scattering time scales as $N^{-1/d}$, i.e., $N^{-0.33}$ for a 3D system and $N^{-0.5}$ for a 2D system. The good agreement between our experimental results and this simple model indicates, that at these densities, carrier-carrier scattering is strongly suppressed by Coulomb screening. As a result of this screening, each carrier effectively interacts only with the distribution of nearest-neighboring carriers.

This interpretation of the power law observed in Fig. 3 requires several assumptions. The first assumption is that excitons play a minor role in our observations. Two facts confirm this hypothesis. First, on our time scale and for the highest densities, excitons are ionized [18]. Second, the population of excitons created with the large bandwidth of the short pulse is small compared to the 2D free-carrier component as seen in Fig. 1. Our results are also consistent with previous measurements [19] of 2D excitonic dephasing times performed on a GaAs single quantum well. Honold *et al.* have shown that the contribution of excitons to the dephasing process occurs on the picosecond time scale. Moreover, in their experiment, the broadening of the homogeneous excitonic linewidth varies linearly with the exciton density. Therefore, the effect of excitons on screening is different from that of a 2D free-carrier gas, which confirms that the $N^{-0.55}$ power law reported here is not due to excitons.

Another question which arises from our results concerns the distribution of carriers in k space. As seen in Fig. 1, the pulse spectrum covers a very broad range of energies. However, this will not substantially effect the Coulomb screening by nearest neighbors, since the Coulomb interaction will occur even between carriers with different energies. The distribution of carriers in k space may affect the magnitude of the dephasing times but not their power-law dependence, in which we are interested here.

Another assumption made in our ballistic interpretation of the two-dimensional screening is that electrons and holes play the same role in the dephasing process. As mentioned earlier, the degree of confinement for electrons and heavy and light holes is different for $n=1$ and $n=2$ subbands. Therefore, it is very likely that the different carriers contribute with different weights to the $N^{-0.5}$ power law. An attempt to experimentally investigate the different contributions of $n=1$ and $n=2$ subbands by reducing the pulse bandwidth was not successful. By reducing the pulse bandwidth, we found that the dephasing process was hardly resolved. This effect might be due to the diminishing temporal resolution and to the relative increase of excitonic population (which contributes differently to the screening, as reported in Ref. [19]) with respect to the 2D free-carrier component. Instead of reducing the pulse bandwidth, one might obtain information on the different carrier contributions to the screening by investigating several doped quantum wells. Nevertheless, the simple nearest-neighbor interaction picture gives a fairly good description of our measurements. Naturally, a complete theoretical description of our experimental results requires sophisticated many-body calculations taking into account the highly nonthermal carrier population created by the femtosecond pulses.

In conclusion, we have reported the first observation of a two-dimensional screening behavior in the dephasing process which occurs between an electron and a hole created by an interband optical transition using 8-fs infrared pulses. We find that the decay time during which the electron and the hole maintain phase coherence is proportional to $N^{-0.55}$, where N is the carrier density. We attribute this behavior to a strong Coulomb screening which is due to the polarization cloud created by the other carriers. We are able to explain our results, as well as previous measurements performed in a three-dimensional system, by assuming a short-range interaction due to Coulomb screening with an effective screening length which scales to the radius of a disk containing the nearest neighbors of a given carrier.

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[1] E. P. Wigner, Phys. Rev. **46**, 1002 (1934).

[2] For a recent review on ultrafast spectroscopy of semiconductors, see E. O. Göbel, in *Feskörperprobleme, Advances in Solid State Physics,* edited by V. Rössler (Vieweg, Braunschweig, 1990), Vol. 30, p. 269.

[3] J. Shah, IEEE J. Quantum Electron. **22**, 1728 (1985).

[4] P. C. Becker, H. L. Fragnito, C. H. Brito-Cruz, R. L. Fork, J. E. Cunningham, J. E. Henry, and C. V. Shank, Phys. Rev. Lett. **61**, 1647 (1988).

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

[6] M. T. Portella, J.-Y. Bigot, R. W. Schoenlein, and C. V. Shank (to be published).

[7] C. V. Shank, R. L. Fork, R. Yen, J. Shah, B. I. Greene, A. C. Gossard, and C. Weisbuch, Solid State Commun. **47**, 981 (1983).

[8] W. H. Knox, C. Hirlimann, D. A. B. Miller, J. Shah, D. S. Chemla, and C. V. Shank, Phys. Rev. Lett. **56**, 1191 (1986).

[9] W. H. Knox, D. S. Chemla, G. Livescu, J. E. Cunningham, and J. E. Henry, Phys. Rev. Lett. **61**, 1290 (1988).

[10] S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).

[11] L. Schultheis, J. Kuhl, A. Honold, and C. W. Tu, Phys. Rev. Lett. **57**, 1635 (1986); **57**, 1797 (1986).

[12] C. V. Shank, P. C. Becker, H. L. Fragnito, and R. L. Fork, *Ultrafast Phenomena VI* (Springer-Verlag, Berlin, 1988), p. 344.

[13] P. C. Becker, H. L. Fragnito, R. L. Fork, F. A. Beisser, and C. V. Shank, Appl. Phys. Lett. **54**, 411 (1989).

[14] R. L. Fork, C. H. Brito-Cruz, P. C. Becker, and C. V. Shank, Opt. Lett. **12**, 483 (1987).

[15] This configuration is referred to as the self-diffraction photon echo as opposed to the stimulated photon echo as used, for instance, by Noll *et al.* to measure excitonic dephasing times in CdSSe mixed crystals: G. Noll, U. Siegner, S. Shevel, and E. O. Göbel, Phys. Rev. Lett. **64**, 792 (1990).

[16] A review on this type of calculations is given in G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Editions de Physique, Paris, 1988), Chap. 3.

[17] See, for instance, D. Pines and C. Nozieres, *The Theory of Quantum Liquids: Normal Fermi Liquids* (Addison-Wesley, Reading, MA, 1966).

[18] D. S. Chemla, Helv. Phys. Acta **56**, 607 (1983).

[19] A. Honold, L. Schultheis, J. Kuhl, and C. W. Tu, Phys. Rev. B **40**, 6442 (1989).