Carrier dephasing in the gain region of an inverted semiconductor

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The polarization dephasing time has been measured across the gain region, at the transparency point, and into the absorption region of an optically excited GaAs multiple-quantum-well sample using spectral hole burning and degenerate four-wave mixing techniques. We observe strongly energy-dependent dephasing rates with a minimum at the crossover from gain to absorption. Numerical results for the microscopically calculated carrier-carrier scattering rates for a two-component electron-hole plasma in quantum wells show fair agreement with the experimental findings.

As originally predicted by Luttinger,[1] in a degenerate one-component electron plasma at zero temperature carrier-carrier scattering rates are expected to vanish at the chemical potential,[2] and carrier–LO-phonon scattering rates should be zero in a region of $2\hbar\omega_{LO}$ around the Fermi energy.[3] Similar expectations hold for a two-component electron-hole (e-h) plasma in highly excited semiconductors for both bulk [three-dimensional (3D)] and quantum-well (quasi-2D) structures. Investigation of the microscopic properties of electron-hole plasmas is important for both basic physical understanding as well as application-oriented studies of semiconductor diode amplifiers and lasers.[4,5]

An experimental investigation of scattering rates in a degenerate electron plasma obtained by n-modulation-doped multiple quantum wells was recently reported.[6] However, the Fermi edge was always near the band edge, so that the dephasing time was not clearly seen to decrease between the Fermi edge and the band edge. In this paper, we report experiments for a two-component electron-hole plasma which demonstrate a definite decrease of the scattering rate at the transparency point. This decrease of the scattering rate manifests itself as a lengthening of the polarization time or the T_2 time for the carriers.

Due to the large carrier density introduced by our optical pumping, we investigate the carrier dephasing well below and above the Fermi energy for a dense electron-hole plasma in a semiconductor at low temperatures. Optical excitation by a strong femtosecond pulse situated far above the band edge prepares a large spectral region of optical gain [see Fig. 1(a)] extending many meV below the transparency point (TP). To measure polarization dephasing rates in this strongly inhomogeneously broadened region, both spectral hole burning (SHB) and degenerate time-integrated four-wave mixing (TI-FWM) techniques were used. In the SHB technique, a spectral hole is burned in the gain region of the semiconductor with its width being related to the T_2 time.[7] The width of the spectral hole and its spectral position in the gain region varies as the pump frequency is changed, thus allowing the measurement of the frequency-dependent T_2 time. This technique is most appropriate for short T_2 times where the hole spectral width is much larger than the pump pulse linewidth. For long T_2 time measurements, we employ the TI-FWM technique in two-beam self-diffraction geometry. In this method, the excitation energies are kept within the $\chi^{(3)}$ limit,[8] and the dephasing

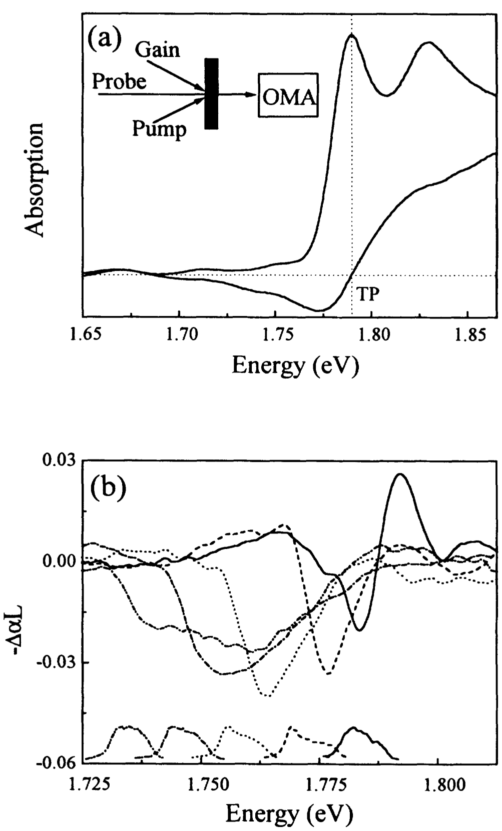

FIG. 1. (a) Linear absorption (upper curve) and absorption 12 ps after the gain pulse (lower curve). The gain pulse at 2 eV excites $\approx 1.8\times10^{12}$-cm^{-2} carriers. Inset: The experimental arrangement. (b) Differential absorption for several pump energies with the effect of probe chirp numerically removed. The pump delay time for each curve is -50 fs, and the pulse energy is 50 nJ. From left to right on the bottom, the pump photon energies are 1.734, 1.747, 1.759, 1.771, and 1.784 eV.

time is obtained from an $\exp(-T_2/4)$ fit to the decay of the correlation trace.[9,10] The spectral and temporal measurements are complementary, and both are needed in order to measure dephasing times on the order of a pulse width. When one technique loses resolution, the other becomes useful.

To perform these experiments, three beams are required from our laser system: (1) a "gain" beam used to excite the sample; (2) a weak broadband probe beam for the SHB experiment; and (3) a tunable, narrow-band pump beam for both SHB and TI-FWM. To generate these beams, we utilize a femtosecond laser system based on a colliding-pulse mode-locked (CPM) laser. The 2.0-eV pulses produced by the CPM are amplified in a six-pass, bow-tie, dye amplifier pumped by a frequency-doubled, diode-pumped Nd:YLF (yttrium lithium fluoride) laser running at 1 kHz. A small portion of this amplified 2.0-eV light is split off and sent to the sample as the "gain" beam. The majority is used to create a broadband continuum. A small portion of this continuum is split off and serves as the broadband probe in the SHB experiment. The rest of the continuum goes through an interference filter, and is reamplified in a second six-pass dye amplifier pumped by a second Nd:YLF laser. This beam is tuned by tilting an interference filter and serves as the pump beam in both experiments as well as the probe in the TI-FWM. For the interference filters used, the temporal width is approximately 150 fs.

The samples used for this work are molecular-beam-epitaxially grown multiple quantum wells consisting of 150 periods of 26-Å GaAs wells and 80-Å $Al_xGa_{1-x}As$ ($x=0.41$) barriers. The sample used for TI-FWM has an antireflection (AR) coating for 685 nm on one surface. For the SHB experiment, the sample is AR coated on both surfaces in order to further reduce the Fabry-Pérot fringes. The samples are kept in a cryostat at approximately 20 K.

The SHB results are shown in Fig. 1. Figure 1(a) shows the linear absorption (upper curve) and absorption 12 ps after the arrival of the gain pulse (lower curve). The gain region extends from about 1.682 eV to the transparency point in the 1.78–1.79-eV range. There is no reliable way to determine the actual temperature of the optically injected plasma or the carrier density accurately. The only experimentally known quantity is the spectral width of the gain region, which can be used to determine the injected carrier density. From this measured gain width, we estimate a carrier density of 1.8×10^{12} cm^{-2}. The inset to Fig. 1(a) illustrates the experimental setup. Differential absorption at several pump photon energies and the corresponding pump pulse spectra are shown in Fig. 1(b). In all curves, the pump delay is fixed at -50 fs in order to probe carriers before too many have a chance to scatter. A spectral hole in the gain is seen at each pump energy.[7] For pump energies in the gain region, the hole is negative, indicating decreased transmission due to depletion of the gain. A blueshift of the spectral hole with respect to the pump energy is also evident. This was found to be caused primarily by a reduction in the band-gap renormalization due to carrier loss. At 1.784 eV, the pump spectrum covers the TP, resulting in a combined positive and negative signal corresponding to depletion of the gain in the region below the TP, and bleaching of the absorption in the region above the TP. At longer-time delays, the spectral hole scatters into a carrier-depletion signal that extends across the gain region and into the absorption region.

The variation of the T_2 as a function of photon energy is clearly evident in Fig. 1(b). At low photon energies, far from the TP, the hole is quite broad, corresponding to rapid dephasing. Near the TP, the hole width is nearly pump limited, indicating a slower dephasing. To determine the T_2 time, the pump spectrum is deconvolved from the spectral hole width, and the resulting width is interpreted as the convolution of two Lorentzian homogeneous linewidths; i.e., $T_2=2/\Delta v\pi$, where Δv is the fre-

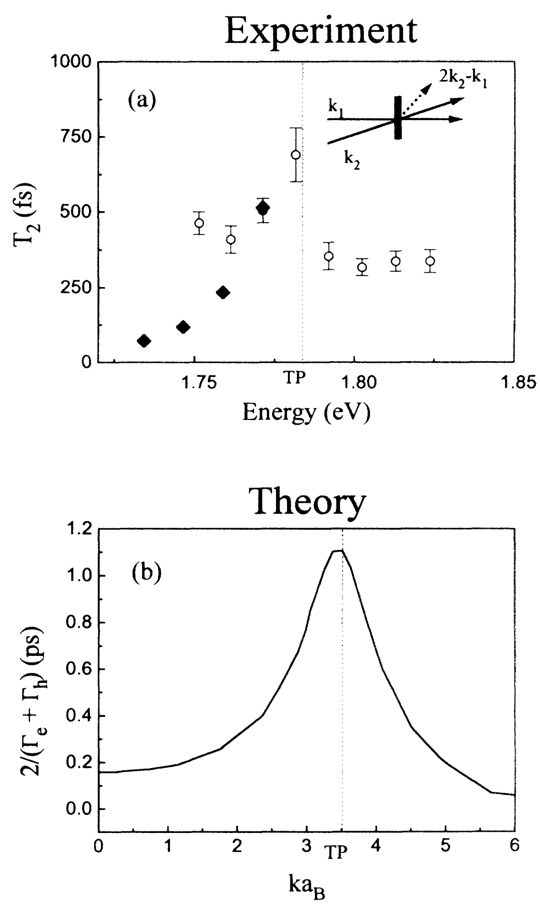

FIG. 2. T_2 times obtained from time-integrated self-diffraction (circles with error bars). The energy of the pulse in direction k_2 varied from 20 nJ in the gain to 0.5 nJ in the absorption region. The energy of the pulse in direction k_1 was an order of magnitude lower. The gain pulse at 2 eV excites $\approx1.8\times10^{12}$-cm^{-2} carriers and arrives 60 ps prior to the mixing pulses. The error bars represent uncertainty in fitting the correlation trace. The filled diamonds are the T_2 times extracted from the spectral hole burning experiment. (b) Calculated dephasing times from carrier-carrier scattering for a plasma temperature of 40 K and a carrier density of 1×10^{12} cm^{-2}. The parameters used are $m_e=0.067m_0$, $m_h=0.107m_0$ (m_0 is the free-electron mass), and background dielectric constant $\varepsilon_b=12.7$. The well width is 26 Å.

quency width of the burned hole. We obtain T_2 times ranging from ≈ 70 fs at 1.734 eV to ≥ 500 fs at 1.771 eV. These T_2 times are shown by the diamond symbols in Fig. 2(a).

Figure 2(a) also shows the time domain results from the TI-FWM measurements. In this experiment, the energy of the pulse in direction k_1 is an order of magnitude lower than that in direction k_2, and the pulses are orthogonally polarized. The pulses for this experiment have spectral full width at half maximum (FWHM) of approximately 8.9 meV for photon energies <1.775 eV, and 10.0 meV for photon energies >1.775 eV. The data points from this measurement are shown by the circles, and were obtained by fitting the TI-FWM signal as previously described.[9,10] In this experiment it is possible to measure both far above and below the TP. Away from the TP on both sides, the measured T_2 is limited by the laser temporal pulsewidth. However, near the TP, we find T_2 significantly longer: 700 fs. Within their respective resolution limits, the data from SHB and TI-FWM are in far agreement.

The theoretical explanation of the above effect can be based on the arguments given by Luttinger in 1960.[1] Recent quantitative studies for carrier-carrier scattering rates in electron-hole plasma have been performed for bulk semiconductors.[2,11] We have extended these calculations for a two-component electron-hole plasma for a semiconductor quantum well, similar to Ref. 12. The theory is a quasiequilibrium evaluation of the so-called Lennard-Balescu equation:

$$\left.\frac{df_\alpha(k_1)}{dt}\right|_{C\text{-}C} = \Gamma^\alpha_{\text{in}}(k_1,f)[1-f_\alpha(k_1)] - \Gamma^\alpha_{\text{out}}(k_1,f)f_\alpha(k_1)\ ,$$

where $\alpha = e$ for electrons, or h for holes. The Γ's are the in and out scattering rates defined as

$$\Gamma^\alpha_{\text{in}}(k_1,f) = \frac{2\pi}{\hbar}\sum_{\alpha',k_2,k_3,k_4} 2|W[k_2-k_1,\varepsilon_\alpha(k_2)-\varepsilon_\alpha(k_1)]|^2 f_\alpha(k_2)[1-f_{\alpha'}(k_3)]f_{\alpha'}(k_4) \times \delta_{k_1+k_3,k_2+k_4}\delta[\varepsilon_\alpha(k_1)-\varepsilon_\alpha(k_2)+\varepsilon_{\alpha'}(k_3)-\varepsilon_{\alpha'}(k_4)]\ .$$

Γ_{out} is obtained from Γ_{in} by replacing f by $(1-f)$. $\varepsilon(k)$ are the parabolic one-particle energies, and W is the dynamically screened Coulomb potential, where we use the screening function in the random-phase approximation. In quasithermal equilibrium the distribution functions f are Fermi functions, and the k-dependent dephasing rate for the optical probe polarization is

$$\frac{1}{T_2} = \tfrac{1}{2}\{\Gamma^e_{\text{in}}[k_1,f_{\text{Fermi}}] + \Gamma^e_{\text{out}}[k_1,f_{\text{Fermi}}] + \Gamma^h_{\text{in}}[k_1,f_{\text{Fermi}}] + \Gamma^h_{\text{out}}[k_1,f_{\text{Fermi}}]\}\ .$$

Here we have neglected intervalence-band and intersubband scattering, restricting ourselves to a description of very thin quantum wells. The Coulomb potential contains the usual form factor for the case of infinite barrier potentials. Since we are dealing with an inverted semiconductor where the interband Coulomb correlation plays only a minor role, we also neglect polarization scattering effects (i.e., dephasing contributions of off-diagonal elements in $\mathbf{k}$). Using parameters for 2D GaAs, we obtain the total scattering rate $(\Gamma_e+\Gamma_h)$ as a function of plasma temperature at the Fermi edge for three different carrier densities, as shown in Fig. 3. For each density, the scattering rate increases with rising plasma temperature and the corresponding dephasing time decreases. The rate at a given temperature decreases as density increases because the plasma becomes more degenerate. These calculations can be used to obtain the dephasing times for electrons and holes due only to carrier-carrier scattering for any plasma temperature. The result of such a calculation for a plasma temperature of 40 K, which is representative of our experiment, is shown in Fig. 2(b). A clear maximum is observed at the Fermi edge, and the times decrease to either side.

The experimental results in Fig. 2(a) can be compared with the calculated dephasing times of Fig. 2(b). Qualitatively the same behavior is seen: a large dephasing time at the TP which then decreases for spectral regions away from the TP. Exact quantitative comparison between experiment and theory is problematic for several reasons. First, the reduced carrier-carrier scattering increases the relative importance of additional scattering processes at the TP. These are mainly carrier–LO-phonon scattering, impurity scattering, surface roughness, and other scattering processes. These processes give rise, as an example, to the zero-density exciton linewidth in bulk material. However in a strongly inhomogeneously broadened sys-

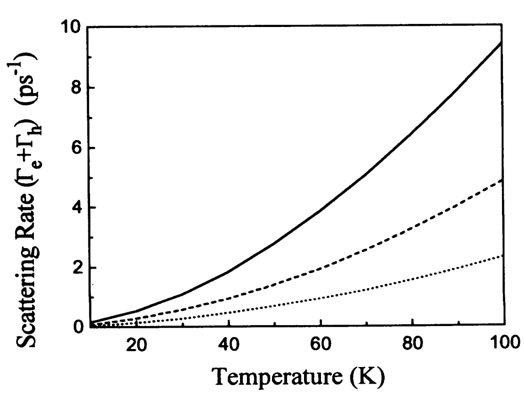

FIG. 3. Calculated scattering rates at the Fermi energy as a function of temperature for carrier densities of 1×10^{12} (solid curve), 2×10^{12} (dashed curve), and 4×10^{12} cm^{-2} (dotted curve).

tem like quantum wells, this homogeneous zero-density linewidth cannot easily be determined, and a theoretical computation of these effects is not within the scope of this investigation. The carrier–LO-phonon scattering rates, on the other hand, can be neglected at low temperatures in a region of $\pm\hbar\omega_{LO}$ around the chemical potential, which is roughly the experimentally accessible regime. Second, it is difficult to make an exact experimental determination of the carrier density and plasma temperature. For this reason, calculated plots like Fig. 3 for the temperature- and density-dependent carrier-carrier scattering rate at the Fermi wave vector are very useful. In principle, these data allow us to follow the carrier cooling processes, since, for a given density, the scattering rate at $\mathbf{k}_F$ decreases monotonically with temperature. Finally, another factor affecting the maximum measured T_2 time comes from the measurement technique. Experimentally, dephasing times are averaged across the spectral width of the pulse. Theoretically, scattering rates are calculated for individual energies. This will cause the measured peak to fall below the calculated peak.

The behavior of the T_2 time near the transparency point is a consequence of a carrier distribution that is almost entirely occupied below the chemical potential, and almost entirely empty above it.[1] Since every carrier-carrier scattering event requires two carriers (electrons and/or holes) to exchange and conserve both energy and momentum, there are very few such events that can happen to a test charge near a sharp Fermi edge. The total scattering rate is therefore low near the TP and rises as we move away from the TP. In this description, the sharpness of the distribution at the Fermi edge provides the key to the behavior of the scattering rate. For a step function (0 K), the scattering rate at the Fermi edge will theoretically go to zero. As the edge becomes less sharp ($T>0$), some states above the Fermi energy are occupied and some states below the Fermi energy are vacant. Thus the scattering rate is nonzero at the Fermi energy and becomes larger as the Fermi edge is more smeared out.

In conclusion, we have used spectral hole burning and four-wave mixing to observe long dephasing times near the transparency point, and shorter dephasing times on either side of it in an optically inverted semiconductor. Within their respective limits of resolution, the spectral and temporal methods yield very similar results. The results agree qualitatively with our theoretical work on carrier-carrier scattering by a cold plasma.

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