Ultrafast Electron Redistribution through Coulomb Scattering in Undoped GaAs: Experiment and Theory

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We report the observation of spectral hole burning exclusively due to the nonequilibrium electron population in a nondegenerate pump-test configuration. The rapid redistribution of electrons as well as the other features of the differential absorption spectra are well described by a theory using quantum-kinetic bare Coulomb collisions in the framework of the semiconductor Bloch equations. [S0031-9007(96)01740-1]

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The redistribution of nonequilibrium carrier populations in semiconductors has attracted considerable interest in the last two decades. The tremendous progress of femtosecond lasers in terms of pulse duration and stability has rendered possible the observation of the initial stages of carrier relaxation [1–6] and the study of very low carrier densities [6]. However, studying the contributions of different scattering mechanisms such as LO-phonon and carrier-carrier scattering remains a difficult task, because most experiments measure a combination of electron and hole dynamics and the signals in ultrashort-pulse experiments contain coherence effects [7] and are not solely population dependent. Indeed, standard pump-test experiments [1,3,5,6] measure the absorption saturation due to the Pauli exclusion principle and are sensitive to the sum of the electron and hole distribution functions (f_e and f_h, respectively) while time-resolved luminescence experiments [2] measure the product $f_e f_h$. A selective investigation of the hole dynamics has been used in Ref. [8] to measure the heavy-hole thermalization time. However, this method cannot measure the complete hole distribution and the initially injected hole population.

Here we have used a modified pump-test scheme in order to isolate the electron dynamics [4]: the pump pulse excites electrons from the heavy-hole (HH) and light-hole (LH) valence bands while the test pulse probes the absorption saturation of the interband transition from the split-off (SO) valence band to the conduction band C (see inset of Fig. 1). Because of the large spin-orbit splitting in GaAs (340 meV), no holes are present in the SO band and the differential absorption signal $-\Delta\alpha = \alpha_{\text{without-pump}} - \alpha_{\text{with-pump}}$ depends on the electron distribution only. This method has a further important advantage: pump and test are at different wavelengths which allows the observation of spectral hole burning due to the initially injected electron population without any contribution from the induced-grating coherence effect [7,9] which considerably complicates the interpretation of standard pump-test experiments. Moreover, due to the isotropic matrix element of the SO-C transition, the measured signal is equally sensitive to the presence of electrons with all possible wave vector directions.

We report the first observation of hole burning which can be attributed exclusively to the electron population. While in previous experiments hole burning was not discernible [4], recent ameliorations of the experimental setup have permitted the observation of hole-burning signals for carrier densities ranging from a few 10^{15} to a few 10^{18} cm^{-3} and for excess photon energies ranging from 50 to 110 meV. The ensemble of these results will be discussed elsewhere. In this Letter, we concentrate on the very short pump-test delay times at moderate densities, the rapid redistribution of electrons causing the

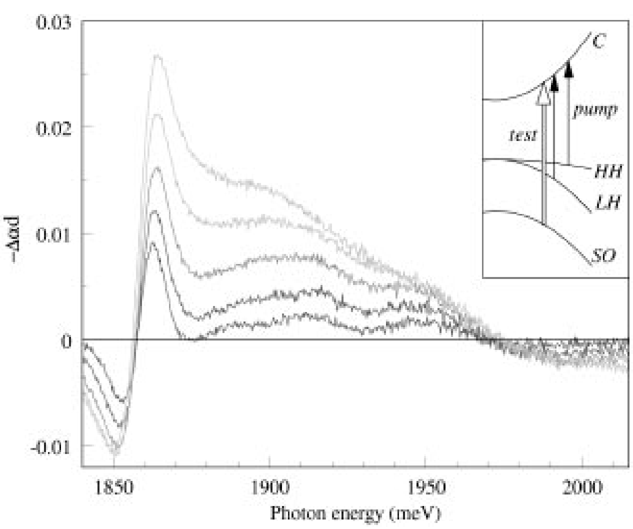

FIG. 1. Differential absorption spectra for the following pump-test delay times: −80, −40, 0, 40, and 80 fs. The inset shows the pump-test configuration.

disappearance of hole burning, and the comparison of this early-time behavior with theory.

We used a Ti:sapphire mode-locked oscillator (Coherent Mira) and regenerative amplifier system (Coherent RegA) both pumped by an argon-ion laser. Part of the output generates a spectral continuum and is used as the test pulse. After chirp compensation of the continuum with a combination of prisms and gratings we obtain nearly Fourier-transform-limited pulses with a duration of 30 and 130 fs for the test and pump pulses, respectively. The pulses were focused down to 50 and 150 μm for the test and pump, respectively, on the sample which was an intrinsic GaAs layer of thickness $d = 0.65$ μm antireflection coated on both sides and held at 15 K. In order to minimize the noise, a shutter is used in the optical path of the pump at an 8-Hz rate. In addition, a reference beam is simultaneously detected on a different track of the CCD detector and is used to normalize the transmitted test beam.

The differential absorption spectra for a pump width of 15 meV and a pump energy of 1.589 eV (excess energy of 70 meV with respect to the band gap) and for various pump-test delay times are shown in Fig. 1. The carrier density was estimated to be 6×10^{16} cm^{-3}. The zero delay is defined as the coincidence of the pump and test maxima and is taken at the middle of the integrated-signal rise time. The spectra show two broad peaks at about 1.913 and 1.950 eV due to spectral hole burning associated with the electron populations photoexcited from the LH and HH bands, respectively. Note that the two peaks disappear already before the end of the pump pulse. While it is clear that the signal in the spectral region from 1.88 to 1.96 eV is dominated by the induced transmission due to the electron population, the induced absorption above 1.97 eV and the oscillatory structure around 1.86 eV cannot be easily explained. Furthermore, even in the hole-burning region the differential absorption spectra do not directly reflect $f_e(t)$ due to energy-time uncertainty and excitonic Coulomb effects. Therefore, a theoretical analysis in terms of a quantum-kinetic approach is necessary, since the commonly used theories based on the golden-rule long-time limit are not applicable at such ultrashort times.

Quantum kinetics is a generic name for the theory describing kinetics with memory on very short time scales (see Ref. [10] for a review). The Markovian rate equation which has been so successful in the description of picosecond and nanosecond phenomena should be regarded as a limiting case of the quantum kinetics. In the experimental results described in this Letter, many scattering mechanisms are involved. It is most interesting to look at the limited short-time regime where Coulomb scattering dominates because, although the quantum kinetics of the electron-LO-phonon interaction (at low carrier densities) has already received attention in the past few years [11–15] and some observed quantum-kinetic effects have been explained [16], no treatment of the quantum-kinetic Coulomb scattering for real experiments has been attempted yet.

Coulomb scattering presents peculiar features which require imperatively a quantum-kinetic formulation. It is well known already from the equilibrium theory of screening that the screened Coulomb potential has an $\frac{1}{q^2}$ singularity as $q \to 0$ at any finite frequency ω. The singularity is absent only at $\omega = 0$. However, a vanishing frequency implies an infinite time. Therefore, the singularity is always present and plays an important role at short time scales. This singularity which corresponds to that of the bare Coulomb potential is fatal for the Boltzmann equation since the argument of the energy-conserving δ function also vanishes at $q = 0$ and the collision integral diverges. The energy-time uncertainty which is taken into account by quantum kinetics automatically eliminates the divergence [17].

For times less than a typical plasma period, screening is negligible [17] and thus the relatively complicated theory of time-dependent screening [18–20] can be avoided. We may also simplify the theoretical task by restricting our calculations to times less than or comparable with the effective interband polarization decay time. In our configuration, where there is no interference between the pump and test polarizations, one can use a simple phenomenological description of the polarization collision term and concentrate only on the quantum-kinetic collision terms of the electron and hole populations excited by the pump.

The semiconductor Bloch equations [21] for the populations and polarizations in the case of the pump field are

$$\frac{\partial f_{e,\vec{k}}(t)}{\partial t} = \sum_{\alpha} \Im\{\Omega^{P*}_{\alpha,\vec{k}}(t) p_{\alpha,\vec{k}}(t)\} + \frac{\partial f_{e,\vec{k}}(t)}{\partial t}\bigg|_{\text{coll}} \quad (1)$$

$$\frac{\partial f_{\alpha,\vec{k}}(t)}{\partial t} = \Im\{\Omega^{P*}_{\alpha,\vec{k}}(t) p_{\alpha,\vec{k}}(t)\} + \frac{\partial f_{\alpha,\vec{k}}(t)}{\partial t}\bigg|_{\text{coll}} \quad (2)$$

$$\left[\frac{\partial}{\partial t} + \frac{i}{\hbar}(\epsilon_{e,\vec{k}} + \epsilon_{\alpha,\vec{k}} - \hbar\omega_P)\right] p_{\alpha,\vec{k}}(t) = \frac{i}{2}\Omega^{P}_{\alpha,\vec{k}}(t)(1 - f_{e,\vec{k}} - f_{\alpha,\vec{k}}) + \frac{\partial p_{\alpha,\vec{k}}(t)}{\partial t}\bigg|_{\text{coll}} . \quad (3)$$

Here $\alpha = \text{HH}, \text{LH}$ and the renormalized energies ϵ and Rabi frequencies Ω are given by

$$\epsilon_{i,\vec{k}}(t) = \epsilon^0_{i,\vec{k}} - \sum_{\vec{k}'} V_{\vec{k}-\vec{k}'} f_{i,\vec{k}'}(t), \qquad i = e, \text{HH}, \text{LH}, \quad (4)$$

$$\hbar\Omega^P_{\alpha,\vec{k}}(t) = d_{\alpha,\vec{k}}\mathcal{E}_P(t) + 2\sum_{\vec{k}'} V_{\vec{k}-\vec{k}'} p_{\alpha,\vec{k}'}(t) . \quad (5)$$

In the above equations, $\mathcal{E}_P(t)$ is the envelope of the pump field with frequency ω_P, $d_{\alpha,\vec{k}}$ are the respective interband dipole matrix elements, and $V_{\vec{q}}$ is the Fourier transform of the Coulomb potential. Since we consider an isotropic model with dipole matrix elements independent of the field polarization and $\vec{k}$, we take them to be equal for heavy and light holes.

The quantum-kinetic collision terms for the populations are ($i, i' = e$, HH, LH):

$$\frac{\partial f_{i,\vec{k}}(t)}{\partial t}\bigg|_{\rm coll} = -\frac{4}{\hbar^2}\sum_{i'}\int_{-\infty}^{t} dt' \int \frac{d\vec{q}}{(2\pi)^3}\int \frac{d\vec{k}'}{(2\pi)^3}|V(q)|^2 \cos\left(\frac{(t-t')}{\hbar}(\epsilon^0_{i,\vec{k}}+\epsilon^0_{i',\vec{k}'}-\epsilon^0_{i,\vec{k}-q}-\epsilon^0_{i',\vec{k}'+q})\right) \times \{f_{i,\vec{k}}(t')f_{i',\vec{k}'}(t')[1-f_{i,\vec{k}-\vec{q}}(t')][1-f_{i',\vec{k}'+\vec{q}}(t')] - f_{i,\vec{k}-\vec{q}}(t')f_{i',\vec{k}'+\vec{q}}(t')[1-f_{i,\vec{k}}(t')][1-f_{i',\vec{k}'}(t')]\}. \quad (6)$$

In the Markovian limit one gets from this equation the usual golden-rule rate equation.

The phenomenological collision term of the polarization is

$$\frac{\partial p_{\alpha,\vec{k}}(t)}{\partial t}\bigg|_{\rm coll} = -\frac{1}{T_2}p_{\alpha,\vec{k}}(t), \qquad \alpha = \text{HH, LH}. \quad (7)$$

In the case of the test pulse, one may retain only the electron population created by the pump and, therefore, we have to consider only the test polarization equation

$$\left[\frac{\partial}{\partial t} + \frac{i}{\hbar}(\epsilon_{e,\vec{k}} + \epsilon^0_{\mathrm{SO},\vec{k}} - \hbar\omega_T)\right]p_{\mathrm{SO},\vec{k}}(t) = \frac{i}{2}\Omega^T_{\mathrm{SO},\vec{k}}(t)(1-f_{e,\vec{k}}) - \frac{p_{\mathrm{SO},\vec{k}}(t)}{T_2}, \quad (8)$$

where the unrenormalized energy of the SO holes $\epsilon^0_{\mathrm{SO},\vec{k}}$ and the renormalized SO Rabi frequency were introduced,

$$\hbar\Omega^T_{\mathrm{SO},\vec{k}}(t) = d_{\mathrm{SO},\vec{k}}\mathcal{E}_T(t) + 2\sum_{\vec{k}'}V_{\vec{k}-\vec{k}'}p_{\mathrm{SO},\vec{k}'}(t). \quad (9)$$

Here $\mathcal{E}_T$ is the envelope of the test field having the carrier frequency ω_T and $d_{\mathrm{SO},\vec{k}}$ the SO dipole matrix element. To obtain the absorption spectrum, one has to perform a Fourier transform of the test polarization summed over all $\vec{k}$.

The electron population excited by the pump acts first as a final-state blocking factor on the right hand side of Eq. (8) and second as a band shift through the Fock energy of the renormalized electron energies [Eq. (4)]. These effects are all mixed up, vary in time, and get Fourier transformed and therefore it is very difficult to discuss them separately. In addition, specific Coulomb spectral effects of the Wannier operator in the polarization equation (exciton and Coulomb enhancement) impede a simple additive interpretation.

Using a 130-fs pump pulse we performed calculations of the excited populations up to 300 fs which corresponds roughly to the plasma period at our pair density of 6×10^{16} cm^{-3}. We took $T_2 = 130$ fs. The effective mass ratios were taken to be integer ($m_{\rm HH}/m_e = 6$, $m_{\rm LH}/m_e = 1$, $m_{\rm SO}/m_e = 2$) for convenience of the numerical algorithm. The numerical calculation on a discrete lattice of k-space points neglects low-momentum-transfer contributions which in the Coulomb case are important. Nevertheless, we take into account low-momentum-transfer collisions within a Landau approximation through a Taylor expansion around $\vec{q} = 0$. The complete quantum-kinetic calculation gives rise to an electron population as shown in Fig. 2. The electron-population peaks are rapidly smeared out and already at about 300 fs after the pump maximum the distribution is very close to a nondegenerate Fermi distribution. The calculated differential absorption spectra with a 30-fs test pulse are shown in Fig. 3. We did not consider delay times longer than 80 fs since their calculation involves information on the electron population for times above 300 fs due to the Fourier transform.

The agreement with the experiment is surprisingly good, although in many details quantitatively rough. The most remarkable achievement is the prediction that at about 80 fs after the pump maximum the induced hole burning is smeared out. This is related to the fact that the electron population is almost in equilibrium already at about 300 fs after the pump maximum. The only fit parameter was the phenomenological polarization relaxation time T_2. However, if T_2 is taken comparable or larger than the pump duration, it affects only the negative parts of the spectra slightly. In the comparison of theory and experiment, one has to take into account that the exact energy positions of the various features are affected by the roughness of the electron energy discretization of about 5 meV, by the slightly modified effective masses as well as by the inaccuracy of the numerical Fourier transform. The more pronounced valley above the band threshold (at about 15 meV) as compared to the experiment may be due to the neglect of LO-phonon emission, which provides the cooling of the electron system.

Both the experimental and the theoretical differential absorptions show a final-state occupation effect (hole burning) due to our narrow-band excitation and an oscillation

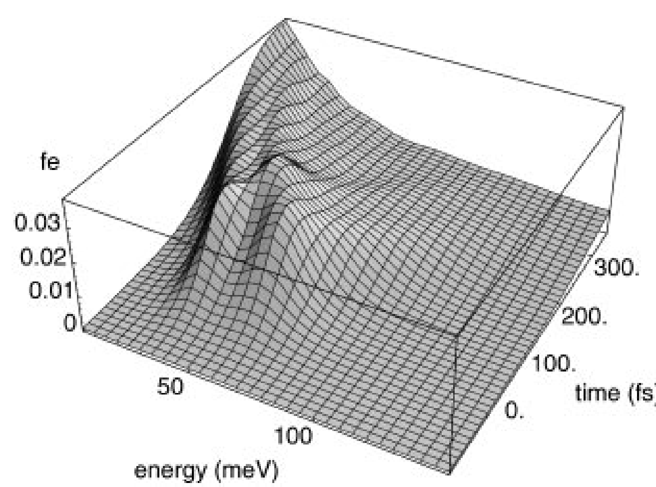

FIG. 2. Quantum-kinetic evolution of the electron population.

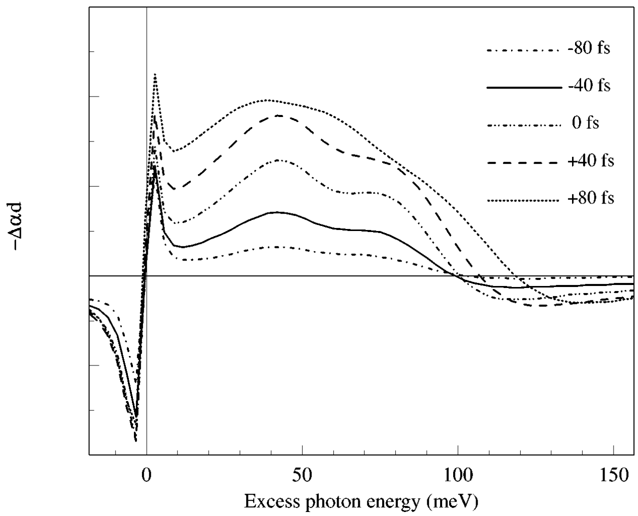

FIG. 3. Calculated differential absorption spectra for the same delay times as in Fig. 1.

that looks like an energy shift in the excitonic region, while the negative signal on the high-energy side of the excitation is mainly due to the nonlinearity introduced by the product of the occupation factor with the Coulomb force term

$$\frac{i}{\hbar}\sum_{\vec{k}'} V_{\vec{k}-\vec{k}'} p_{\mathrm{SO},\vec{k}'}(t)\left[1 - f_{e,\vec{k}}(t)\right], \tag{10}$$

which stems from the replacement of the Rabi frequency by the renormalized one in the presence of the Coulomb interaction [see Eq. (9)]. Actually, this excitonic enhancement term plays an important role also in the oscillatory structure on the low-energy side in addition to the true Coulomb band shift of Eq. (4).

In conclusion, we have observed hole burning in a pump-test configuration free from coherence effects and succeeded in giving a satisfactory description of the differential absorption spectra for ultrashort delay times with the semiconductor Bloch equations using the quantum-kinetic bare Coulomb collision term for the populations. We stress that the use of quantum kinetics is mandatory due to the Coulomb singularity. The main success of the theory is determined by the structure of the semiconductor Bloch equations but the numerical prediction of the effective intraband relaxation time (smearing out of the hole burning) is due entirely to quantum kinetics. Our theoretical approach was highly simplified due to the specific experimental configuration implying a differential signal determined only by the electron population.

An improved version of the theory should include the quantum-kinetic polarization collision term, the buildup of screening, LO-phonon collisions, as well as the transition to the Markovian behavior in order to extend its applicability to higher densities and longer times. To incorporate such improvements in the theory for the test beam will still be an insurmountable task due to the Fourier transform, which requires an enormous time interval.

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