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PHOTOCARRIER RELAXATION IN THE QUANTUM KINETICS REGIME

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Abstract

We present a study of carrier relaxation in GaAs in the Quantum-Kinetics regime. The results cannot be explained by Boltzmann Kinetics. They are in satisfactory agreement with Quantum-Kinetic theory calculation of the occupation number in k-space.

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The thermalization of nonequilibrium carrier distributions involve fundamentals of thermodynamics and solid states physics. Femtosecond spectroscopy has been employed to study this process,[1] and was analysed in the framework of Boltzmann Kinetics.[2] However, during and shortly after an ultrashort laser excitation, carrier kinetics are drastically altered. At this early stage the Coulomb potential is still unscreened and there is essentially no energy conservation in two-particle scattering events, instead scattering becomes non-local in time. In this regime the evolution of momentum-dependent occupation numbers can only be described by Quantum Kinetics, including memory structure in the scattering integrals.[3]

We present a study of carrier relaxation in bulk GaAs at room temperature in the Quantum-Kinetics regime. We use a modified frequency-resolved pump-probe technique, with pump and probe durations independently adjustable from 30fs to 100fs.

Figure 1a shows differential absorption spectra (DAS) for 70fs pump and probe at low excess energy. The evolution from a hole-burning-like signal with a characteristic Dynamic Fermi-Edge Singularity (DFES)[4,5] at $\Delta t \sim -50$ fs, to a quasi-thermalized profile at $\Delta t \sim 30$ fs is clearly seen, in agreement with previous experiments[1]. DAS obtained with 30fs pulses are shown in Fig. 1b, the higher excitation energy minimizing excitonic effects. Surprisingly, the DAS show an instantaneous response at low energies. Within ~ 50 fs the DAS is characteristic of a thermalized distribution. The negative signal on the high-energy side, characteristic of the DFES, is absent. There is practically no response at the peak of the pump spectrum, in fact *hole-burning is not observed at all*. Such a behavior cannot be explained by Boltzmann Kinetics, which predict gradual diffusion of the initial spectral hole. With 100-fs excitation

high above the band edge and a 30-fs probe we also detect an instantaneous response at low energies and no spectral hole (Fig. 2). This last result is in agreement with another recent experiment.[6]

These observations cannot be explained in terms of coherent effects. Screening by nonequilibrium carriers is expected to be ineffective, as indicated by the excitons' response. It should not reduce the Sommerfeld enhancement significantly. Emission of optical phonons is clearly important. However, evaluation of the emission rate by Fermi golden-rule is inappropriate, since the inverse optical phonon energy is comparable to the time scale of the measurements.[7]

A consistent explanation can only be given in the framework of Quantum Kinetics. A complete model of the DAS in this formalism is presently unavailable. However, we give, in Fig. 3, an example Quantum Kinetic calculation that includes electron-electron scattering only, showing the time-evolution of the occupation number in k-space for excitation by a 20-fs pump pulse well above the band edge. The photocarriers rapidly scatter out of the energy window in which they were created. Due to the memory, kinetic overshoots develop. We find indications of such overshoots also in the experimental data. A complete description of the DAS requires inclusion of the short coherent probe. We stress that a very short probe is *essential* for the observation of Quantum Kinetic effects, which are due to the non-locality of scattering. for a probe longer than the range of non-locality (memory depth), >70 fs, the kinetics become indistinguishable from local Boltzmann Kinetics.

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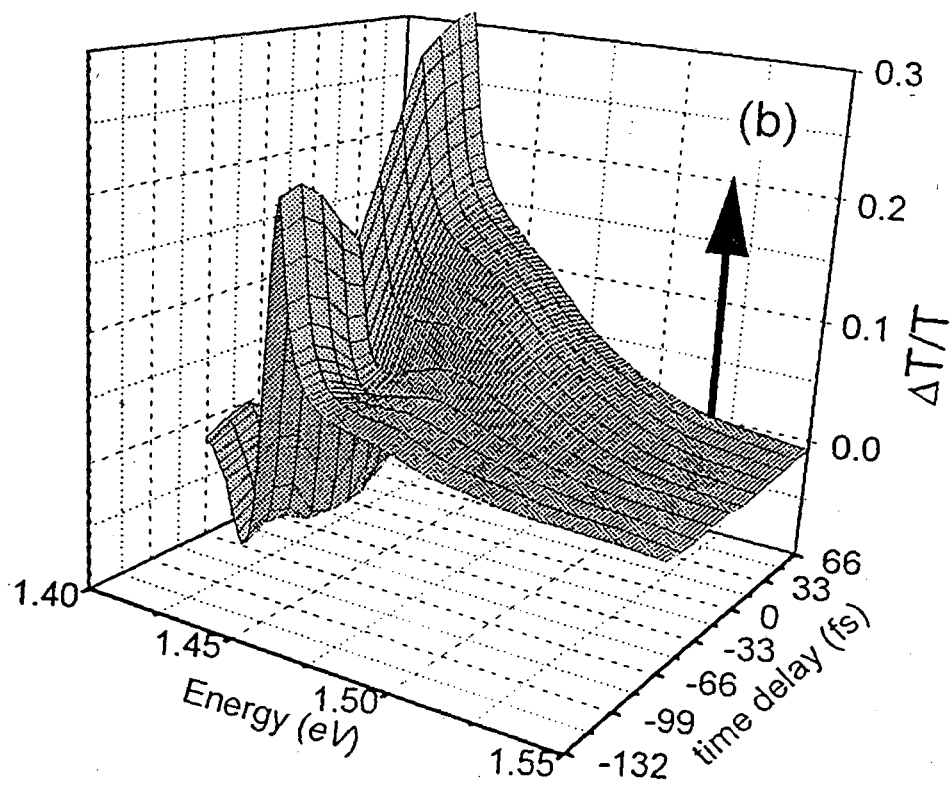
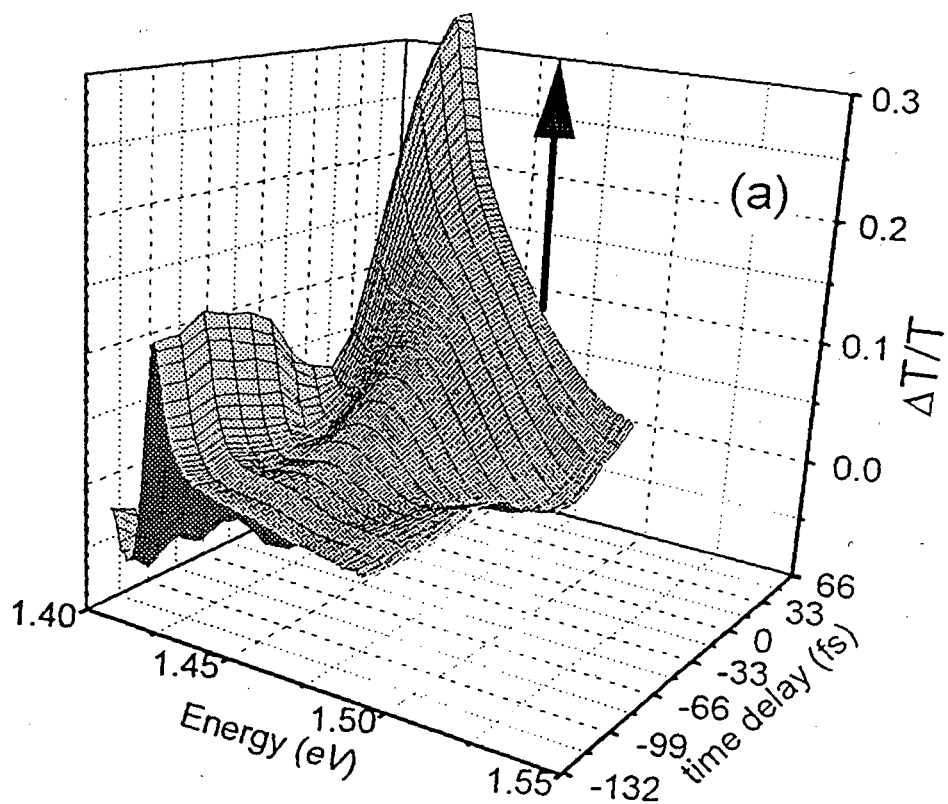
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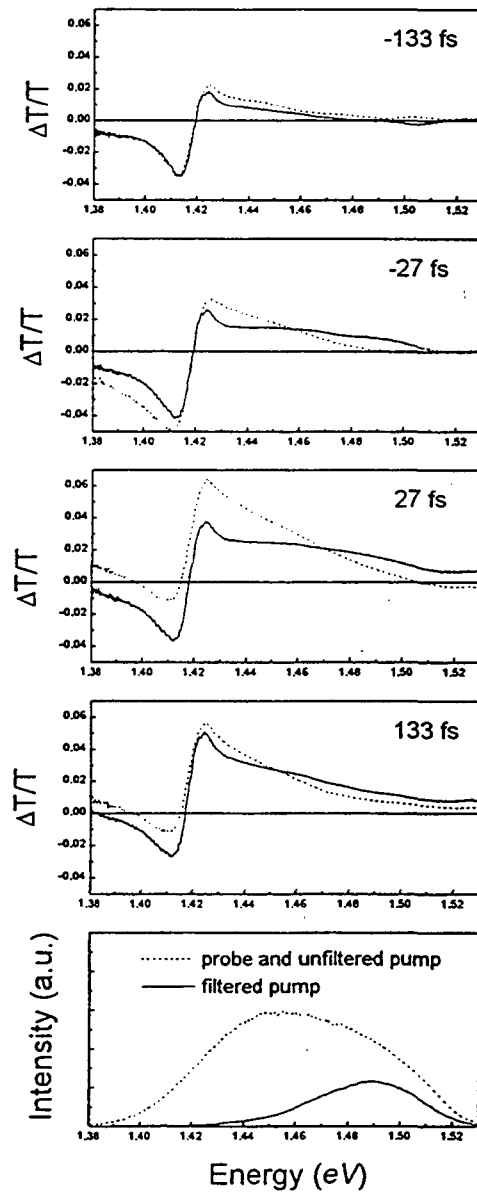
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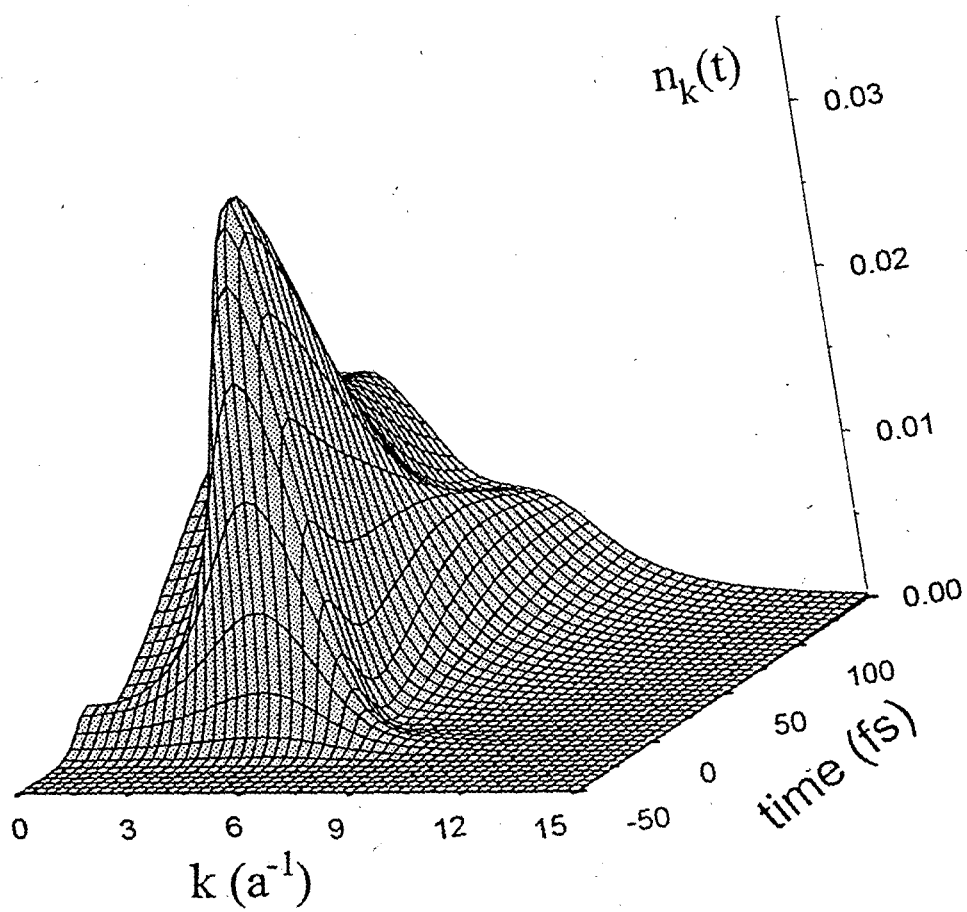
Figure 1: Spectrally-resolved differential absorption signals measured with 70-fs pulses (a) and 30-fs pulses (b). The signals, measured for a carrier density of $\sim 1 \cdot 10^{17} \text{ cm}^{-3}$, are representative of lower excitation densities as well, down to $\sim 1 \cdot 10^{16} \text{ cm}^{-3}$. The zero delay is identical for both experiments. The arrows indicate the center laser energy in each case.

Figure 2: Spectrally-resolved differential absorption signals measured with a 30-fs probe and two different pump durations - 100 fs (solid) and 30 fs (dashed). In both cases an immediate response is observed at the band edge. The bottom panel shows the spectrum of the 100-fs pump (solid) and that of the 30-fs probe/unfiltered pump (dashed).

Figure 3: The time-evolution of the occupation number in k-space after an excitation with a single 20 fs pump pulse, calculated in the framework of Quantum Kinetic theory.







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