Relaxation dynamics of electrons between Landau levels in GaAs

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The relaxation dynamics of free electrons between Landau levels is traced on a picosecond time scale via the time evolution of the band-to-acceptor luminescence in p-type GaAs after resonant excitation of the $n=1$ Landau level. For electron densities $N>10^{14}$ cm^{-3}, relaxation occurs via electron-electron scattering with a $1/e$ decay time of 7.5×10^{15} ps cm$^{-3}/N$. For lower electron densities, relaxation becomes independent of density due to elastic impurity scattering. [S0163-1829(96)52524-5]

A strong magnetic field imposed on a semiconductor splits the energy band of free electrons and holes into a series of quantized Landau levels, separated by the cyclotron energy $\hbar\omega_c$. The splitting can be varied easily by tuning the magnetic field B. Each Landau level is actually a one-dimensional band since carrier motion is free along the direction of the magnetic field. Transitions between Landau levels can therefore be used for tunable far-infrared (FIR) sources or detectors.[1] Therefore, detailed knowledge of the mechanisms and dynamics of relaxation between Landau levels is desirable.

Saturation absorption[2–5] and induced conductivity changes[6,7] of cyclotron resonance have been used to study relaxation between Landau levels. However, most of these methods are time-integrated measurements. They require the solution of a model with coupled rate equations in order to determine the interlevel relaxation times. The time-resolved measurements of conductivity changes[6,7] provide more direct access, but their interpretation is rather complicated and ambiguous. Time-resolved photoluminescence of the band-to-band recombination has been performed;[8] however, carrier density was in this experiment so high that immediately, within the time resolution, a thermal distribution between the Landau levels is established.

We use time-resolved photoluminescence spectroscopy to get a direct insight into the relaxation dynamics of free electrons between Landau levels: ultrashort laser pulses with an excitation energy tuned to the difference between the $n=1$ Landau level of the electron and the heavy hole excite electrons directly into the $n=1$ level. Thus, for a short time, an inversion between the $n=1$ and $n=0$ Landau levels is achieved. Interlevel relaxation is then studied by tracing the time evolution of the band-to-acceptor luminescence with picosecond time resolution. Extremely low excitation densities ($\leqslant10^{15}$ cm^{-3}) are necessary in order to get interlevel relaxation on a picosecond time scale, where highly sensitive detection using a streak camera is possible.[9]

The sample used in our experiment is GaAs:Be grown by molecular-beam epitaxy on semiinsulation GaAs. The room-temperature hole concentration is 6.6×10^{16} cm^{-3}. The sample is kept at 10 K on the cold finger of a He cryostat. We excite with picosecond pulses of a mode-locked Ti:sapphire laser with 80 MHz repetition rate. The photoluminescence is dispersed in a 0.3-m spectrometer and detected by a synchroscan streak camera. Temporal and spectral resolution are 10 ps and 1 meV, respectively.

Figure 1 shows the time evolution of the photoluminescence with resonant excitation of the $n=1$ Landau level at $t=0$. The excitation density is 1.3×10^{14} cm^{-3}. The position of the excitation energy minus the 28 meV acceptor binding-energy[10] is indicated by an arrow. In the spectrum at $t=0$, the band-to-acceptor luminescence of the $n=1$ Landau level (luminescence maximum at 1.506 eV) dominates, revealing a strong overpopulation of the $n=1$ Landau band. In the following 100 ps this overpopulation is reduced via interlevel relaxation until thermal occupation of the Landau bands is reached. The luminescence signal appearing at 1.515 eV at later times is due to the decay of bound excitons and is not relevant for the results discussed here. The dashed lines give the line shapes expected for thermal energy distributions. Very low excitation densities are used in our experiment. The electron density $N(E)$ can therefore be described by a Maxwell-Boltzmann distribution, given by

$$N(E)\propto e^{-E/k_BT}D_B(E), \tag{1}$$

where $D_B(E)$ is the density of states in a magnetic field, according to[11,12]

$$D_B(E) \propto \sum_{n_e=0}^{\infty} \sqrt{\frac{E-(n+\frac{1}{2})\hbar\omega_c+\{[E-(n+\frac{1}{2})\hbar\omega_c]^2+\Gamma^2\}^{1/2}}{[E-(n+\frac{1}{2})\hbar\omega_c]^2+\Gamma^2}}. \tag{2}$$

The broadening factor Γ is determined by the mean electron scattering time τ_e with $\Gamma=\hbar/2\tau_e$.[12] At our low densities, Γ is very small and is assumed to be $\leqslant0.1$ meV (Ref. 11). This broadening is therefore much smaller than the broadening due to the finite width of the acceptor band. The latter is taken into account by additionally convoluting Eq. (1) with a broadening function with a 2.6-meV full width at half maximum, as determined by fitting at later times, when the electron distribution is completely thermal. It is clear that

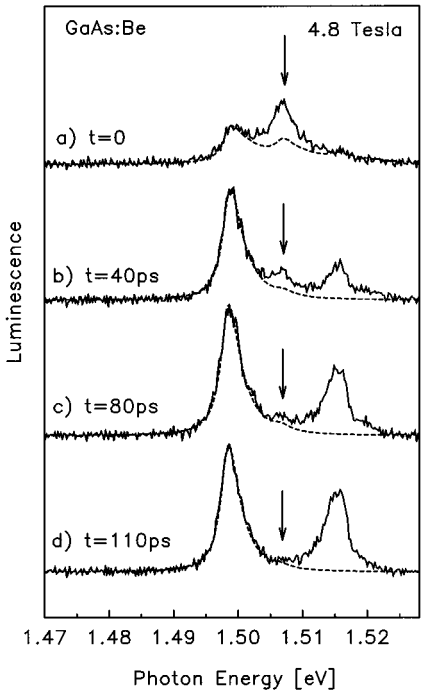

FIG. 1. Transient luminescence spectra of GaAs:Be at 10 K and 4.8 T: (a) 0 ps, (b) 40 ps, (c) 80 ps, and (d) 110 ps after resonant excitation with a short laser pulse into the $n=1$ Landau level. Excitation energy is 1.533 eV with density 1.3×10^{14} cm^{-3}. The luminescence signals with maxima at 1.498 and 1.506 eV are the band-to-acceptor luminescence signals of free electrons in the $n=0$ and the $n=1$ Landau level, respectively. The luminescence at 1.515 eV is due to the decay of bound excitons. The arrow indicates the energy of the laser pulse minus the acceptor energy of Be and corresponds to the band-to-acceptor luminescence of the $n=1$ Landau level. Dashed lines: line-shape fits with Maxwell-Boltzmann distributions convoluted with a broadening function of 2.6 meV full width at half maximum. The temperatures obtained by the fits are (a)$>$100 K, (b) 29 K, (c) 26 K, and (d) 23 K. The relative accuracy of the temperatures is for (b)–(d) about 1 K. The cooling is due to acoustical-phonon emission.

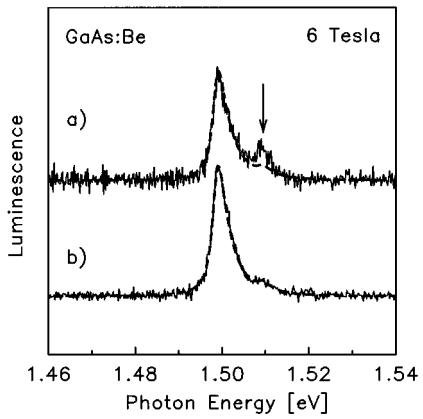

FIG. 2. Band-to-acceptor luminescence of GaAs:Be at 6 T integrated over the first 40 ps after excitation with a short laser pulse: (a) excitation at 1.536 eV, i.e., resonantly into the $n=1$ Landau level, (b) excitation at 1.533 eV, i.e., exciting electrons only in the $n=0$ Landau level. The excitation density is 1×10^{14} cm^{-3}. Dashed lines: spectra for thermal distributions, both with a temperature $T=43$ K. In this figure the bound-exciton luminescence was subtracted for reasons of clarity.

this broadening is the same for all Landau levels and therefore does not affect the relative population of the Landau levels as obtained from the fits.

The fits have then two free parameters: the temperature of the electron gas and the relative occupation of the $n=1$ and $n=0$ Landau levels. However, both parameters are determined independently. The temperature is obtained from the shape of the high-energy side of the recombination involving the individual Landau levels; higher temperature yields a flatter high-energy side, in particular, of the recombination involving the $n=0$ Landau level. The relative occupation of the first two Landau levels is obtained from the relative intensities of the two recombination lines.

Figure 2 shows the luminescence spectra of the band-to-acceptor transitions integrated over the first 40 ps (a) for resonant excitation of the $n=1$ Landau level and (b) for excitation with 3 meV lower energy. The bound-exciton luminescence was subtracted in this figure for reasons of clarity. Carrier-carrier scattering establishes a thermal distribution within the $n=0$ Landau level in a time much faster than our time resolution. Polar optical scattering is for carrier temperatures >50 K, very efficient and also faster than our time resolution. The initial carrier temperature becomes, therefore, independent of the excess energy if it is larger than about 5 meV.[13] In case b, where carriers are excited only into the $n=0$ band, the line shape of the band-to-acceptor luminescence can already be fitted by a thermal distribution at these early times, whereas for resonant excitation of $n=1$ (curve a) the deviation of the observed electron distribution from a Maxwell-Boltzmann distribution clearly reveals an overpopulation of the $n=1$ Landau band.

In order to determine the interband relaxation times, the difference between the experimental band-to-acceptor luminescence spectra and the fits with a thermal distribution is integrated. We cannot use the spectrum at long delay times for subtraction since the temperature still decreases on this time scale due to emission of acoustical phonons. These values are plotted versus time, and the resulting curve is fitted by an exponential decay, directly yielding the relaxation time between Landau levels. The results obtained for various excitation densities are compiled in Fig. 3.

The density dependence shows two regimes: At excitation densities $N_{\rm exc}<10^{14}$ cm^{-3}, interlevel relaxation becomes independent of density, for higher densities the relaxation time varies with $1/N_{\rm exc}$. In a simple approach, the initial depopulation of the $n=1$ level can be described as the solution of the rate equation

$$\frac{dN_1}{dt}=-AN_1^2-BN_1, \tag{3}$$

where $N_1(t)$ is the overpopulation of the higher Landau level $n=1$ with $N_1(t=0)\approx N_{\rm exc}$. The interlevel relaxation-time τ is then defined by

$$N_1(t=\tau)=\frac{1}{e}N_1(t=0). \tag{4}$$

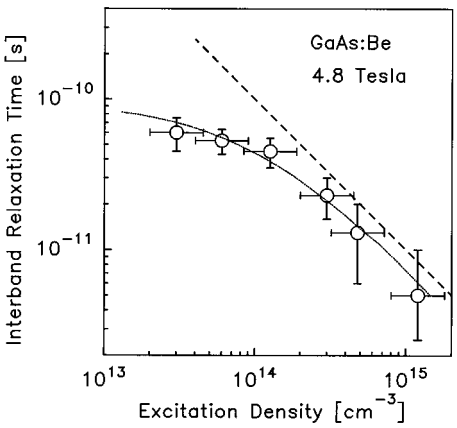

FIG. 3. Intraband relaxation time for the transition from the $n=1$ to the $n=0$ Landau level in GaAs. Circles: experimental data, obtained from the decay time of the integrated difference between the band-to-acceptor luminescence spectra and the fitted spectra assuming thermal distribution. Dotted line: solution of the rate-equation model with $1/A=7.5\times10^{15}$ ps cm^{-3} and $1/B=95$ ps. Dashed line: dependence as calculated for the theoretical estimate for the Auger coefficient with $1/A=1\times10^{16}$ ps cm^{-3}.

The first term on the right side of Eq. (3) mainly determines the depopulation in the regime where τ depends on density,[14] whereas the second term dominates in the regime where τ is independent of density. The coefficients A and B are determined by a fit to the experimental data. The dotted line in Fig. 3 shows the result when the coefficients $1/A=7.5\times10^{15}$ ps cm^{-3} and $1/B=95$ ps are used.

The density-dependent part is caused by an Auger process via electron-electron scattering: one electron scatters from the $n=1$ to the $n=0$ Landau level, the other one from the $n=1$ to the $n=2$ level. Both transitions occur with $\Delta k=0$. Such an Auger process is highly efficient for interband relaxation between Landau levels due to the equidistant Landau splitting.[2,6,15] The efficiency of the Auger process depends on the cyclotron radius and the density. A theoretical estimate of the Auger process is given in Ref. 2 in Eq. (1) and yields a value of $1/A=1\times10^{16}$ ps cm^{-3} for a magnetic field of 4.8 T and a mean energy of about 4 meV. This dependence is shown as a dashed line in Fig. 3.

At excitation densities $N_{\rm exc}<10^{14}$ cm^{-3}, the interlevel relaxation time becomes independent of density. The limiting relaxation time is 95 ps, which is much faster than the values obtained by cyclotron-resonance experiments.[4] Acoustical-phonon scattering or radiative recombination are much too slow to explain this fast relaxation time. However, the relatively high acceptor concentration in our sample can cause efficient elastic impurity scattering, which transfers electrons under energy conservation from the $n=1$ to the $n=0$ Landau level. There the hot electrons thermalize via intraband scattering within our time resolution. The samples used for the cyclotron-resonance experiments[4] had a doping concentration about three orders of magnitude lower than our sample, which explains why we get a much faster lower limit for the interlevel relaxation than Allan *et al.*[4]

In summary, we have for the first time directly traced the dynamics of free-electron relaxation between Landau levels on a picosecond time scale. For densities $>10^{14}$ cm^{-3}, Landau relaxation occurs via an electron-electron Auger process, and for low densities via elastic impurity scattering.

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[1] See, for example, E. Gornik, Physica **127B**, 95 (1984), and references therein.
[2] E. Gornik, T.Y. Chang, T.J. Bridges, V.T. Nguyen, J.D. Mcgee, and W. Müller, Phys. Rev. Lett. **40**, 1152 (1978).
[3] T. Ohyama, J. Phys. Soc. Jpn. **51**, 1431 (1982).
[4] G.R. Allan, A. Black, C.R. Pidgeon, E. Gornik, W. Seidenbusch, and P. Colter, Phys. Rev. B **31**, 3560 (1985).
[5] K. Mitchell, A. Black, C.R. Pidgeon, G.R. Allan, M.F. Kimmitt, and E. Gornik, J. Phys. C **20**, 5217 (1987).
[6] W. Müller, E. Gornik, T.J. Bridges, and T.Y. Chang, Solid State Electron. **21**, 1455 (1978); W. Müller, F. Kohl, and E. Gornik, Infrared Phys. **18**, 691 (1978).
[7] H.J.A. Bluyssen, J.C. Mann, and P. Wyder, Solid State Commun. **31**, 465 (1979).
[8] J.F. Ryan, R.A. Taylor, A.J. Turberfield, and J.M. Worlock, Physica **134B**, 318 (1985).
[9] See also D.W. Snoke, W.W. Rühle, Y.-C. Lu, and E. Bauser, Phys. Rev. B **45**, 10 979 (1992); Phys. Rev. Lett. **68**, 990 (1992).
[10] *Physics of Group IV Elements and III-V Compounds*, edited by O. Madelung, M. Schulz, and H. Weiss, Landolt-Börnstein, New Series, Group III, Vol. 17, Pt. a (Springer Verlag, Berlin 1982).
[11] W. Rühle and E. Göbel, Phys. Status Solidi B **79**, 311 (1976).
[12] L.M. Roth and N. Argyres, in *Semiconductors and Semimetals*, edited by R.K. Willardson, and A.C. Beer (Academic, New York, 1989), Vol. 1, p. 168.
[13] W.W. Rühle, K. Leo, and E. Bauser, Phys. Rev. B **40**, 1756 (1989).
[14] Actually a hyperbolic decay of $N_1(t)$ is expected in this regime. This is indeed experimentally observed at high densities. We approximate, however, the initial decay by an exponential law in order to simplify analysis.
[15] M. Potemski, R. Stepniewski, J.C. Maan, G. Martinez, P. Wyder, and B. Etienne, Phys. Rev. Lett. **66**, 2239 (1991).