Free-electron laser study of the nonlinear magnetophotoconductivity in *n*-GaAs

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The University of California at Santa Barbara free-electron laser was used to investigate the kinetics of electrons bound to shallow donors in *n*-GaAs by saturation spectroscopy. The resonant photothermal conductivity arising from $1s$–$2p^+$ shallow donor excitations in a magnetic field was measured at intensities greatly exceeding that of earlier investigations and saturation of bound-to-free photoionization transitions was achieved. The impurity resonance photoconductive signal shows a distinct intensity dependence caused by competing bound-to-free transitions which saturate differently. This permits a more detailed evaluation of the electron recombination kinetics than was previously possible, yielding the ionization probability of the $2p^+$ state, the transition time of electrons from the $2p^+$ level to the gound state, and the recombination time of free carriers.

At low temperatures, the kinetics of electrons bound to shallow impurities in high-purity semiconductors may be studied by saturation spectroscopy using high-power, far-infrared (FIR) lasers. Several studies involving resonant impurity transitions and cyclotron resonance were carried out using transverse excited atmosphere (TEA)-CO_2 laser pumped molecular FIR lasers[1–3] and electrically pulsed quasi-cw lasers emitting $\sim 100\ \mu s$ pulses.[4–7] The saturation of the $1s$–$2p^+$ shallow donor transition in *n*-GaAs in an external magnetic field B was studied in absorption[4,5] and in photothermal conductivity[6] as a function of applied FIR laser power. Within the intensity range of quasi-cw lasers, the relaxation rate associated with this transition is adequately described in analogy to a two-level system with a single decay constant τ_{eff}. Since τ_{eff} is longer than the pulse width of TEA pumped FIR lasers, they are not useful for higher power, steady-state saturation behavior studies.

In the present work, we examine the saturation behavior of the photothermal conductivity of both the $1s$–$2p^+$ (resonant) and $1s$–free carrier (background) transitions in *n*-GaAs using $1\ \mu s$ FIR pulses from the University of California at Santa Barbara Free Electron Laser (UCSB FEL).[8] The FIR pulses are long enough to obtain steady-state conditions during irradiation and high enough in power to extend the quasi-cw FIR laser saturation measurements made by a factor of 500 in intensity. This made possible background saturation measurements previously unachievable with quasi-cw FIR lasers. Most important, it revealed that the resonant transition relaxation is not describable in terms of a two-level system at higher powers.

The sample used was a high-purity *n*-GaAs epitaxial layer of $14.6\ \mu m$ thickness having an effective donor concentration $P_a = N_d - N_a = 8.3\times 10^{13}\ cm^{-3}$, a compensation ratio $N_a/N_d = 0.75$, where N_a and N_d are the concentrations of acceptors and donors respectively, and mobility $\mu = 1.14\times 10^5\ cm^2/V\,s$ at $T = 77$ K. The FEL was tuned to $\lambda = 164\ \mu m$ in order to complement previously taken data with a quasi-cw CH_3OH laser. At this wavelength the $1s$–$2p^+$ resonance occurs at $B = 2.05$ T with the $2p^+$ level slightly above the $N = 0$ Landau level (Fig. 1). The photoconductive signal G at the center of the resonance was measured over more than four orders of magnitude of FIR intensity I. Over this range of I we found $G(I)$ on resonance cannot be modeled by a two-level system with a single saturation intensity. Instead, it is essential to include excitations both from the $1s$ state directly into the continuum and also into a short-lived metastable state[9] which overlaps the $1s$–$2p^+$ resonance at $B = 2.05$ T.

We analyzed the on resonance data using a three-state model depicted in the inset of Fig. 1. The rate equations are

$$\begin{aligned} \frac{dn}{dt} &= \sigma_c F n_d + X_2^s n_d^* - \tau_1^{-1} n\left(1 + \frac{n}{N_a}\right), \\ \frac{dn_d^*}{dt} &= \sigma_r F(n_d - n_d^*) - (X_2^s + \tau_2^{-1}) n_d^*, \end{aligned} \tag{1}$$

where n, n_d, n_d^*, are the concentrations of electrons in the conduction band, the $1s$ ground state, and the $2p^+$ state, respectively. The absorption cross sections of the $1s$–free carrier and $1s$–$2p^+$ transitions are σ_c and σ_r respectively. F is the photon flux density $I/\hbar\omega$ of the circular polarization of FIR causing each transition. Each transition couples to opposite circular polarizations, although the FIR impinging onto the sample was unpolarized. τ_2^{-1} is the rate at which electrons relax from the $2p^+$ state to the ground state while remaining bound to the donor and X_2^s is the rate at which the electrons get ionized from the $2p^+$ level. τ_1^{-1} is the rate at which free carriers relax to the $1s$ ground state, not the lifetime of the free carriers. The stimulated emission contribution to dn/dt is neglected both because of the large density of continuum states and because the electrons in the conduction band relax to the bottom of the band on a picosecond time scale.[10]

The rate equations were solved under steady-state conditions $(d/dt = 0)$ assuming local neutrality

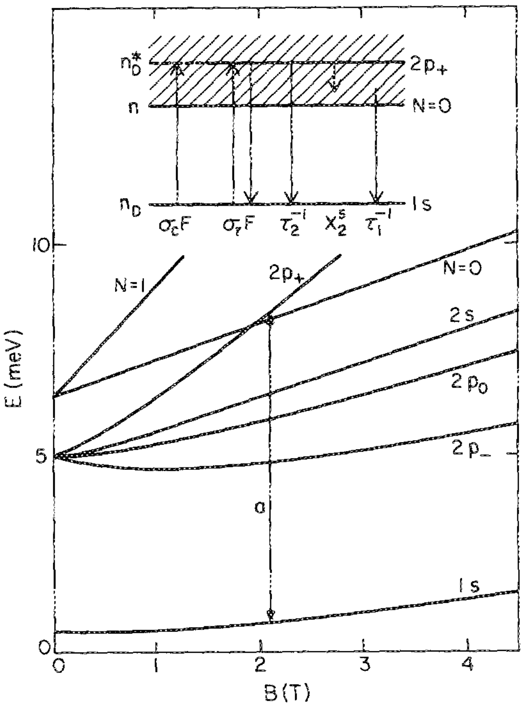

FIG. 1. Shallow donor energy-level splittings of n-GaAs in applied magnetic field. a is the FEL photon energy. Inset is schematic energy-level diagram.

$P_a = N_d - N_a = n_d + n_d^* + n$. We also neglected the n^2 recombination term due to the large number of acceptors ($n/N_a \ll 1$) and because only a small fraction of the available electrons are actually excited to the band. The optically generated free-electron density is found to be

$$n = P_a \tau_1 g, \tag{2}$$

where the generation rate g is

$$g = \frac{p^* \sigma_r F + \sigma_c F + s^* \tau_2 \sigma_c \sigma_r F^2}{1 + 2\tau_{\text{eff}} \sigma_r F + \tau_1 \sigma_c F + \tau_1 s^* \tau_2 \sigma_c \sigma_r F^2} \tag{3}$$

and $p^* = X_2^s/(X_2^s + \tau_2^{-1})$ and $s^* = 1 - p^*$ are the ionization and sticking probabilities of the $2p^+$ level. The effective lifetime is defined as[4]

$$\tau_{\text{eff}} = \frac{X_2^s + 2\tau_1^{-1}}{2\tau_1^{-1}(X_2^s + \tau_2^{-1})} = \frac{1}{2} p^* \tau_1 + s^* \tau_2 . \tag{4}$$

The conductance versus intensity data at three values of B are shown in Fig. 2. All data were taken with the FEL except for the solid circles which were obtained with a quasi-cw laser. The solid curves are fits to $G = e\mu P_a \tau_1 g$ with g given above, where we assume μ is independent of n. The off-resonance data ($B = 1.59$ T and 2.28 T) were fit using g given by Eq. (3) with $\sigma_r = 0$, yielding $n \propto I/(1 + I/I_s)$. This shows that μ is not altered appreciably by saturating the photoionization of shallow donors. On resonance ($B = 2.05$ T) and at the power levels attainable by quasi-cw lasers, the data can be fit using Eq. (3) ignoring the saturation of the background as was done in Ref. 5. At the intensity levels attainable with the FEL it is essential to include the background saturation. The fits to the on-resonance data give a resonant saturation intensity $I_{sr} = \hbar\omega(2\tau_{\text{eff}}\sigma_r)^{-1} = 4.4 \times 10^{-4}\ \hat{I}$ and background saturation intensity $I_{sc} = \hbar\omega(\tau_1\sigma_c)^{-1} = 4.0 \times 10^{-3}\ \hat{I}$, where $\hat{I}$ is the maximum intensity onto the sample of each circular polarization (30 W/cm^{-2}). The background saturation intensity off resonance I_s is greater than I_{sc}, $I_{sc} = 0.11 I_s$. Since τ_1 is not expected to change so rapidly with a small variation in B, we assume the lower value of background saturation intensity on resonance arises from a change in background σ_c.

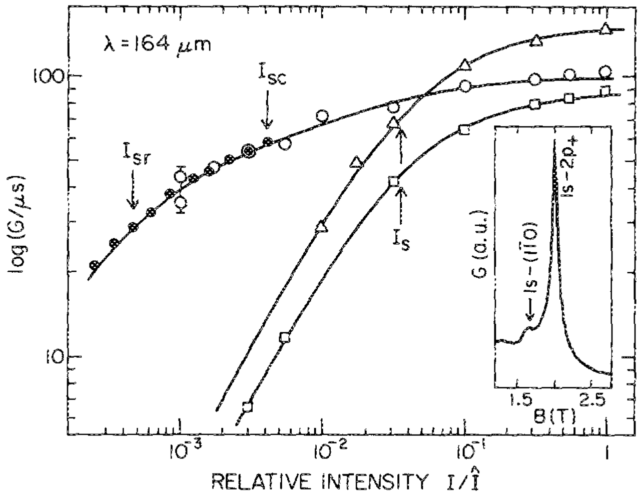

FIG. 2. Photoconductance as a function of intensity in units of peak intensity $\hat{I} = 30$ W/cm^2. Circles: on resonance (2.05 T), full circles are obtained by a quasi-cw laser; triangles and squares: above (2.28 T) and below (1.59 T) resonance, respectively. Saturation intensities are indicated by arrows. Inset is photoconductive signal in vicinity of the $1s$–$2p^+$ resonance. The structure on the low B wing of the line is due to transitions to the (1, − 1,0) metastable donor state.

On the low magnetic field side of the $1s$–$2p^+$ line, a broad structure is observed whose high magnetic field wing overlaps the $1s$–$2p^+$ line (Fig. 2 inset). The energy separation between the peak of this structure and the $1s$–$2p^+$ line is larger than the typical central cell splitting in n-GaAs,[11] thus it cannot be attributed to a different donor. High-resolution Fourier transform spectroscopy revealed that this structure arises from transitions from the donor ground state to an excited metastable state in the $N = 0$ Landau subband with high field quantum numbers $(N,m,j) = (1, -1,0)$.[9] N and m are the Landau level and angular momentum quantum numbers, respectively, and j counts the donor states belonging to one set of N,m. As electrons in the $N = 0$ Landau subband have angular moments $m < 0$, donor states with negative m, in contrast to the $2p^+$ state, are metastable and do not lead to bound states in the $B = 0$ limit. Electrons in such states may freely move into the continuum. Thus, optical excitations of these states simply contribute to the photoionization of donors, although σ is much larger than that of true free-electron states unaffected by the Coulomb potential.[12]

The absorption cross section of this metastable state σ_c (on resonance) could be determined from the ratio I_{sc}/I_s, if σ_c (off resonance) could be measured. Unfortunately σ_c (off resonance) is very small, yielding a small absorbance in the thin epitaxial layers and has not been measured yet. However σ_c (off resonance) can be estimated from theoretical models of the photoionization of shallow donors at large

B. Using the work of Hasegawa and Howard[13] we find σ_c (off resonance) $= 8\times 10^{-15}$ cm^2 for the present experimental conditions. This gives σ_c (on resonance) $= (I_s/I_{cs})\sigma_c$ (off resonance) $= 7\times 10^{-14}$ cm^2, yielding a recombination time $\tau_1 = 144$ ns. From this we obtain the ionization probability of the $2p^+$ level at $B = 2.05$ T to be $p^* = 0.18$ and therefore $\tau_2 = 18.4$ ns. We conclude that only a fraction, possibly as small as 0.18, of the optically populated $2p^+$ levels are actually ionized. This is plausible because the density of states of phonons required to transfer the electron from the $2p^+$ state into the high density of continuum states is very small.

Since τ_1 is the total recombination time, if we let the average capture time of free electrons in the $N = 0$ Landau level be τ_f, then we may write $\tau_1 = \tau_f + \tau_1'$, where τ_1' is the average time the electrons take to cascade down to the ground state. We must then replace Eq. (2) by $n = P_a \tau_f g$ with g still given by Eq. (3). The lifetime τ_f of the present sample at 2.05 T is obtained from the decay of the photocurrent following a FIR pulse which has been switched off[14] $\tau_f = 15$ ns. This gives $\tau_1' = 129$ ns, which is much longer than τ_2. Hence the recombination history of electrons captured in a high donor state below the $N = 0$ Landau level and those excited to the $2p^+$ level and remaining on the donor are very different. It has been argued that the lowest excited state above the ground state, the $2sp$ level in zero magnetic field[15] or the $2p^-$ level in a field,[16,17] represents a bottleneck in electron recombination because of the large energy separation and the small phonon transition probability. Our finding that τ_1' and τ_2 are very different indicates that the electron recombination from the $2p^+$ state cannot be controlled by a long-lived $2p^-$ state. The lifetime of the $2p^-$ has been measured to be 500 ns [5]; hence, there must be a different and faster recombination channel for the $2p^+$ state, probably proceeding through the intermediate $2s$ state.

Under extreme saturating conditions g reduces to τ_1^{-1} yielding the free-electron concentration $n(F\rightarrow\infty) = (\tau_f/\tau_1)P_a = 0.07\ P_a$. Thus, even at high power levels, only a small fraction of available electrons are excited into the conduction band, in keeping with our assumption that the electron mobility remains unaffected under these conditions.

In summary, we have determined the recombination lifetime of electrons in the $N = 0$ Landau level and the lifetime of electrons excited to the $2p^+$ state. Our investigations show that the kinetics of electrons at low temperatures in a high-purity semicondutor may be inferred from saturation measurements using the UCSB FEL. Since the FEL is tunable in the FIR and has sufficiently long pulses, nonlinear spectroscopy need no longer be limited to the few strong molecular laser lines available.

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