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HIGH POWER NON-LINEAR MAGNETOPHOTOCONDUCTIVITY IN n-GaAs USING THE UCSB FREE ELECTRON LASER

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ABSTRACT

The kinetics of electrons bound to shallow donor impurities in n-GaAs was investigated by saturation spectroscopy using the University of California at Santa Barbara free electron laser. The resonant photothermal conductivity from $1s$-$2p^+$ transitions was measured at intensities greatly exceeding previous studies. Saturation of bound-to-free photoionization transitions was measured from 0 to 4 Tesla. The $1s$-$2p^+$ resonant photoconductive signal shows a distinct intensity dependence caused by the competing bound-to-free transitions which saturate differently. Evaluation of the electron recombination kinetics allows us to calculate the transition time of electrons from the $2p^+$ level to the ground state, the recombination time of free electrons, and the thermal ionization probability of the $2p^+$ state.

INTRODUCTION

The kinetics of electrons bound to shallow impurities in high purity semiconductors may be studied, at low temperatures, by saturation spectroscopy using high power, far-infrared (FIR) lasers. Several studies involving resonant impurity transitions and cyclotron resonance were carried out using TEA-CO_2 laser pumped molecular FIR lasers[1,2,3] and electrically pulsed quasi-cw lasers emitting ≈100 μsec pulses[4,5,6,7]. In n-GaAs, the saturation of the $1s$-$2p^+$ shallow donor transition in an external magnetic field

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(B) was studied in absorption[4,5] and in photothermal conductivity[5] 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 by a two level system with a single decay constant, τ_{eff}. Since τ_{eff} is longer than the pulse width of TEA-CO_2 pumped FIR lasers, these lasers are not useful for higher power, steady-state saturation studies.

In this 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 μsec FIR pulses from the University of California at Santa Barbara free electron laser (UCSB FEL)[8]. The FIR pulse width is 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 enabled us to directly measure background saturation intensities, unacheivable with quasi-cw lasers. More importantly, it revealed that the resonant transition relaxation is not describable as a two level system at high powers; instead, it is essential to include both resonant and background excitations in an effective three level model on resonance due to the proximity of the Landau level edge with the $2p^+$ level.

EXPERIMENT

The FEL is a high power source of FIR radiation tunable between 30cm^{-1} to 85 cm^{-1} with approximately 1 kWatt peak power. The time average frequency linewidth of the FEL beam is 1.6 GHz and arises from the fluctuations in the terminal voltage; however the single shot laser beam is single mode with a linewidth of 10 MHz. The FEL was tuned to 164 μm in order to complement previously taken data using a quasi-cw CH_3OH laser and to calibrate the incident FEL power. At this energy, the 1s-$2p^+$ resonance occurs at 2.05 T with the $2p^+$ level slightly higher than the N=0 Landau Level (Fig.1). The photoconductive signal (G) at the center of the resonance was measured over more than 4 orders of magnitude in FIR intensity. In addition, power broadening of the 1s-$2p^+$ transition was observed and background saturation intensities at various magnetic fields between 0T and 4T were measured.

The sample used was a high purity n-GaAs epitaxial layer of 14.6 μm thickness with the substrate bonded to a wedged piece of semi-insulating GaAs to suppress

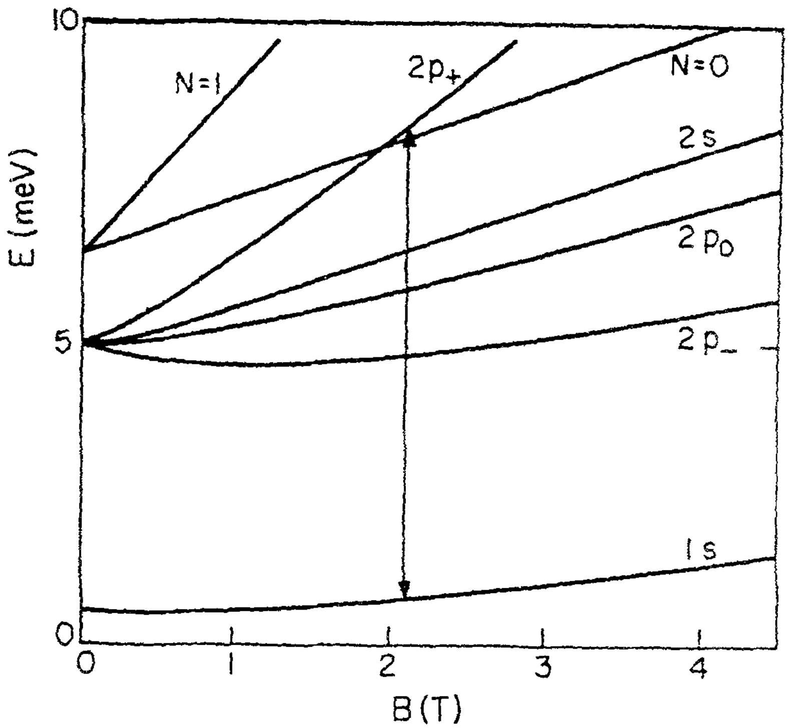

Figure 1. Shallow donor energy level splittings of n-GaAs in an applied magnetic field. Arrow indicates FEL photon energy (164 μm).

interference effects in the sample. The film has an effective donor concentration $P_a = N_d - N_a = 8.3\times10^{13}$ cm^{-3}, a compensation ratio $N_a/N_d = 0.75$, and a mobility $\mu = 1.14\times10^5$ cm^2/Vs. Au-Sn ohmic contacts were deposited on opposite edges.

The magneto-photoconductivity measurements were made in the Faraday configuration using light pipe access with the applied electric field perpendicular to B. The sample was cooled by He exchange gas to 4.2K. Cold crystal quartz and black polyethylene were used as filters to reduce background thermal radiation. Care was taken to assure uniform illumination of the sample. For the quasi-cw CH_3OH laser, a tuned Fabry-Perot interferometer was used to filter unwanted emissions (not 164 μm) from the laser cavity and the intensity was measured with pyroelectric detectors and cross checked with a golay cell. The intensity in the sample was calculated including reflection losses at the front surface and the incident light was unpolarized. A standard bias circuit was employed using a 50 Ohm load resistor to resolve the FEL pulse, and a 500 Ohm load when used with the quasi-cw CH_3OH laser. The bias voltage was always chosen well below breakdown in order to avoid impact ionization nonlinearities.[7,9]

RESONANT TRANSITION MODEL AND RESULTS

The photoconductive signal versus magnetic field is shown in Figure 2 at the lowest power levels using the quasi-cw laser in the vacinity of the $1s\text{-}2p^+$ transition. The signal appears almost Lorentzian and very narrow indicating the predominance of only one chemical species. On the low magnetic field side of the resonance is a broad structure that partially overlaps the $1s\text{-}2p^+$ resonance. This broad resonance occurs at magnetic field strengths greater than 1 T and sharpens with increasing B. The energy separation between the two peaks is larger than the typical central cell splitting in n-GaAs[10] and, therefore, cannot be attributed to a different donor. High resolution Fourier spectroscopy revealed that this broad structure is due to transitions from the donor ground state to an excited metastable state in the N=0 Landau subband with high field quantum numbers $(N,m,j) = (1,-1,0)$[11]. 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. Since electrons occupying the N=0 Landau level have $m \leq 0$, donor states with negative m, in contrast to the $2p^+$ (m=+1) state, are metastable and do not lead to bound states in the B=0 limit. Hence electrons in such states may move freely into the continuum and optical absorption by these states contribute only to the background photoconductivity. The absorption cross section of these states, on the other hand, is much larger than that of the true free electron states[12]. We will estimate this cross-section shortly.

In figure 3 are shown typical magnetic field scans around the $1s\text{-}2p^+$ resonance at different power levels. Notice at high intensity the resonance has disappeared and the background has completely saturated. If we plot the peak photoconductance versus intensity at three magnetic fields: on resonance, above and below (yet well outside) resonance, we see completely different saturation behavior of the resonant and background signals (Figure 4). In fact, the background saturation can be fitted extremely well with a standard two level model, indicating the breakdown of

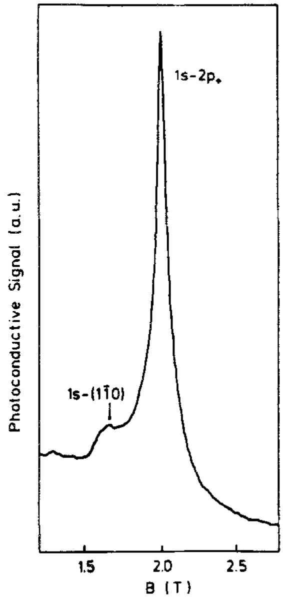

Figure 2. Photoconductive signal versus magnetic field in the vacinity of the 1s-2p$^+$ transition using the quasi-cw laser at low power levels. The structure on the low B side of the resonance arises from transitions to the (1,-1,0) metastable donor state.

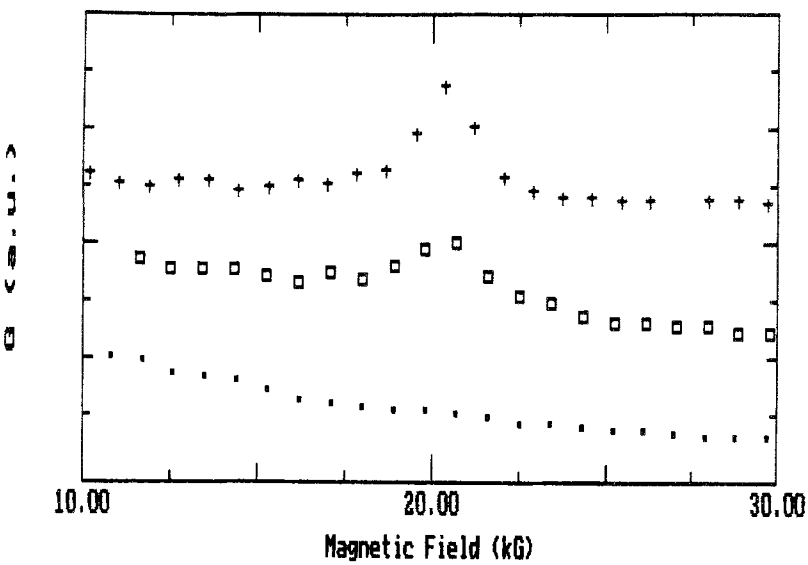

Figure 3. Same as figure 2, except at higher FEL incident power. Solid circles, squares, and crosses are at high, intermediate, and low power levels, respectively.

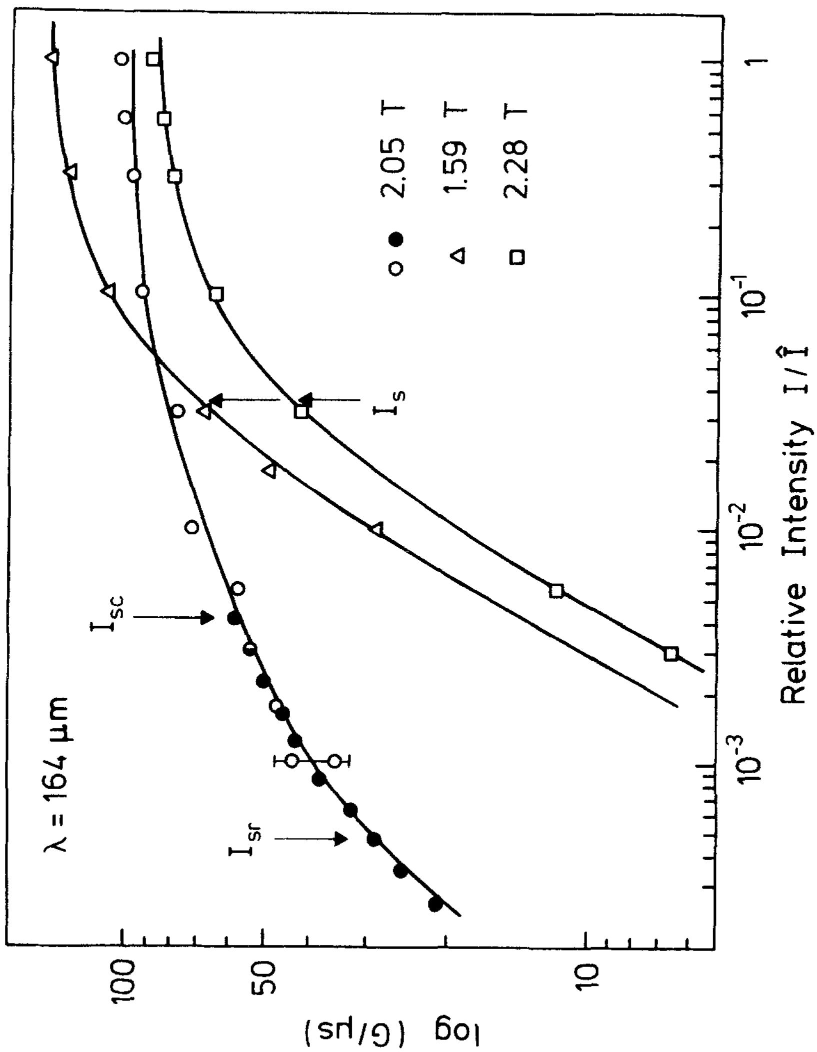

Figure 4. Photoconductance as a function of intensity in units of peak intensity, $I^{\wedge}$ = 30 W/cm^2. Circles: on resonance (2.05 T), full circles are obtained by quasi-cw laser; triangles and square: above (2.28 T) and below (1.59 T) resonance, respectively. Saturation intensities are indicated by arrows.

such a model for the resonance. The low power levels were measured on the quasi-cw laser and matched to give the power levels of the FEL.

The data was analyzed using a three level model with the energy level scheme given in Figure 5. Following this diagram we write the rate equations as:

$$\frac{dn_d^*}{dt} = \sigma_r F(n_d - n_d^*) - (X_2^s + \tau_2^{-1})n_d^*$$

$$\frac{dn}{dt} = \sigma_c F n_d + X_2^s n_d^* - \tau_1^{-1} n(1 + n/N_a) \qquad \text{Eqn. 1}$$

where n, n_d, n_d^* are the concentrations of electrons in the conduction band, the 1s ground state, and the $2p^+$ excited state, respectively. Electrons are excited from the donor ground state directly to the conduction band with transition probability $\sigma_c F$ or to the $2p^+$ state with transition probability $\sigma_r F$. $F = I/\hbar\omega$ with I the intensity of either right or left circular polarization, and σ_c and σ_r are the absorption cross sections of continuum and resonant transitions, respectively. Note that σ_c should contain

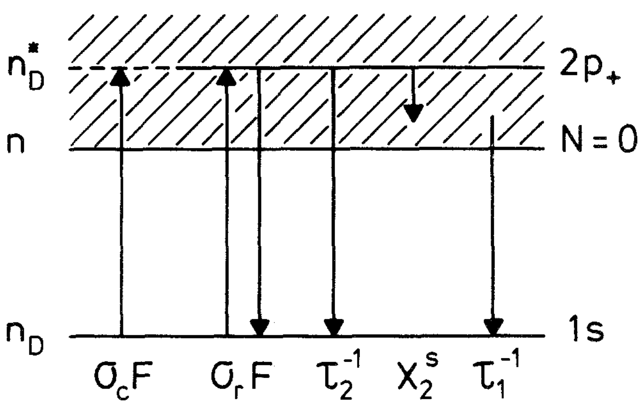

Figure 5. Schematic energy level of n-GaAs used in rate equation model. Symbols are explained in text.

contributions from the metastable state absorption, yet ionization from this state will be much faster than from the $2p^+$ state. Stimulated emission from the free carrier states may be neglected because of the large density of continuum states and because electrons generated at high energies relax to the bottom of the band on a picosecond time scale[13]. Electrons in the $2p^+$ state may ionize into the conduction band with rate X_2^s or stick on the donor and relax to the ground state with time constant τ_2. Free electrons in the band are captured by ionized donors and relax to the ground state within time constant τ_1.

The rate equations are solved under steady state conditions (d/dt = 0) assuming local neutrality $P_a = N_d - N_a = n + n_d + n_d^*$. We neglect the nonlinear n^2 recombination term due to the large number of acceptors ($n/N_a \ll 1$) and, which will be shown, the small fraction available electrons actually excited into the conduction band. The optically generated free electron density is found to be

$$n = Pa\tau_1 g \qquad \text{Eqn. 2}$$

where g, the generation rate, is given by

$$g = \frac{p^*\sigma_r F + \sigma_c F + s^*\tau_2\sigma_c\sigma_r F^2}{1 + 2\tau_{eff}\sigma_r F + \tau_1\sigma_c F + \tau_1 s^* t_2\sigma_c\sigma_r F^2} \qquad \text{Eqn. 3}$$

We have defined $p^* = X_2^s/(X_2^s + \tau_2^{-1})$ and $s^* = 1 - p^*$ as the ionization and sticking probability of the $2p^+$ state. The effective lifetime, τ_{eff} (i.e., the measured low power relaxation time) is given by

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

The measured photoconductance was fit using $G = e\mu n$ assuming the mobility, μ, is independent of n. We will justify this later in the paper.

Let us now examine equation 2 in different limiting and physical cases:

1) Outside Impurity Resonance ($\sigma_r = 0$)

Well outside the $1s$-$2p^+$ resonance our three level model reduces to an effective two level scheme involving just the ground state and continuum with the relaxation time constant τ_1. If we let the density of electrons in the continuum be n_c then outside of the resonance we have

$$n_c = P_a \tau_1 \frac{\sigma_c F}{1 + \tau_1 \sigma_c F} \qquad \text{Eqn. 5}$$

This would give the measured photoconductance, G(I),

$$G(I) = G_\infty \, (I/I_s) \, (1 + I/I_s)^{-1} \qquad \text{Eqn. 6}$$

where G_∞ is the conductance at infinite I, $I_s = \hbar\omega(\tau_1\sigma_c)^{-1}$ as long as the mobility is independant of the number of optically generated free carriers[14].

2) Low Intensity Regime ($I \ll I_s$)

In the intensity range $I \ll I_s$, the photoconductance of continuum transitions depend linearly on I and no saturation behavior is seen, corresponding to the experimental conditions using a quasi-cw laser[5]. If we let $n_r = n - n_c$ be the resonantly generated electron density then,

$$n_r = P_a \tau_1 \frac{\sigma_r F}{1 + 2\tau_{eff}\sigma_r F} \qquad \text{Eqn. 7}$$

This again leads to Equation 6 with saturation intensity, $I_s = \hbar\omega(2\tau_{eff}\sigma_r)^{-1}$.

3) General Case with Unity Ionization Probability ($p^*=1$)

Let us consider when σ_c and σ_r are nonzero yet the probability of the electron leaving the donor is one, $p^*=1$ and $s^*=0$. The total free electron density is then given by

$$n = P_a \tau_1 \frac{(\sigma_c + \sigma_r)F}{1 + (2\tau_{eff}\sigma_r + \tau_1\sigma_c)F} \qquad \text{Eqn. 8}$$

Once again we reproduce Equation 6 with saturation intensity, $I_s=\hbar\omega(2\tau_{eff}\sigma_r+\tau_1\sigma_c)^{-1}$. Referring to Figure 4, since the intensity dependance of G differs drastically on and off resonance we can conclude that only a fraction of the electrons in the optically populated $2p^+$ state are actually excited into the continuum.

The data shown in Figure 4 has been numerically fit to equation 2 where we again assume μ is independent of n.

From the accuracy of the fits we can assume that this is an appropriate assumption. We obtain, on resonance (in units of peak intensity, I_o):

Saturation Intensities(on resonance):

$$I_{sr} = \hbar\omega(2\tau_{eff}\sigma_r)^{-1} = 4.4\text{x}10^{-4} I_o \ ,$$
$$I_{sc} = \hbar\omega(\tau_1\sigma_c(\text{on res}))^{-1} = 4.0\text{x}10^{-3} I_o \ ,$$
where $I_o = 30\ \text{W/cm}^2$.

Saturated Conductance:

$$G_\infty = 98\mu\text{V/A} \ ,$$

thus giving

$$p^*\tau_1 = 0.92\tau_{eff} \ ,$$
$$s^*\tau_2 = \tau_{eff} - p^*\tau_1/2 = 0.54\tau_{eff}$$

Off resonance, $\sigma_r = 0$, we obtain

$$I_s = \hbar\omega(\tau_1\sigma_c(\text{off res}))^{-1} = 3.6\text{x}10^{-2}\ I_o$$

The resonant absorption cross section of the 1s-2p$^+$ transition has been determined from transmission experiments yielding $\sigma_r = 1.8\text{x}10^{-12}$ cm^2 in agreement to previous work[6]. The peak intensity is found to be $I_o = 30$ W/cm^2 of the appropriate polarization. We find that τ_{eff} = 28ns in reasonable agreement with previous saturation data[6].

The saturation intensity I_{sc} of continuum transitions on resonance is much smaller than I_s outside resonance, $I_{sc} = 0.11\ I_s$. Since the recombination time τ_1 is not expected to change with small variations in B, we assume the low value of I_{sc} arises from optical excitations to the (1,-1,0) metastable state. From the ratio I_s/I_{sc} we can determine σ_c(on res) if we could measure σ_c(off res). In the very thin high purity GaAs samples available, the background absorption cross section, off resonance, is very small and until it can be measured we must estimate it on theoretical grounds. Using the work of Hasegawa and Howard[15] on the photoionization of shallow donors in high magnetic fields, we find σ_c(off res)= 8 x 10^{-15} cm^2 for our experimental conditions and hence σ_c(on res)= I_s/I_{sc} x σ_c(off res) = 7 x 10^{-14} cm^2. We can now completely characterize the excitation and relaxation kinetics. The recombination time, τ_1, is found to be 144 nsec, the ionization probability of the 2p$^+$ state, p*, is 0.18, and τ_2 is 18.4 nsec. Interestingly we find that only 18% of excited electrons leave the donor site and conduct even though the $2p^+$ level lies near a high density of continuum states. We might note that although the N=0 density of states is large, the thermal energy necessary for ionization and hence the phonon energy and phonon density of states is 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 $\tau_{(capture)}$ then we may write $\tau_1 = \tau_{(capture)} + \tau_{(cascade)}$ where $\tau_{(cascade)}$ is the average time for electrons to cascade down to the ground state from highly excited donor states. We must then replace τ_1 by $\tau_{(capture)}$ in Eqn. 4 (i.e. $n = P_a \tau_{(capture)} g$), however, in equation 5, τ_1 remains leaving the expression for g unchanged. The lifetime $\tau_{(capture)}$ can be determined at low temperatures by observing the decay of the photocurrent upon fast switching off of the FIR[16]. At 2.05T (on resonance) we measure $\tau_{(capture)}$ = 10 nsec giving $\tau_{(cascade)}$ = 134 nsec which is *much longer* than τ_2. Electrons captured from the band into highly excited donor states obviously have a much different relaxation history than electrons sticking to the donor and returning to the ground state. It has been argued that the lowest excited state above the ground state, the 2sp level in zero magnetic field[17] or the $2p^-$ level in a field[18,19], represents a bottleneck in electron recombination because of the large energy separation and small phonon transition probability. Our result that τ_2 and $\tau_{(cascade)}$ differ by almost an order of magnitude shows that relaxation from the $2p^+$ state cannot be controlled by a long lived $2p^-$ state. If the $2p^-$ state is long lived (>100 nsec) then there must be a different and faster recombination channel for the $2p^+$ state probably through the 2s intermediate state.

To show that the mobility μ remains unaffected at high power level note that under extreme saturating conditions equation 5 gives $g = \tau_1^{-1}$. Hence only a small fraction of available electrons $n_\infty = (\tau_{(capture)}/\tau_1)P_a = 0.07P_a$ are excited into the conduction band even at the highest powers attainable.

POWER BROADENING AND NON-RESONANT TRANSITIONS

In addition to the fixed field resonant data, we were able to study power broadening of the $1s$-$2p^+$ resonant transition as shown in figure 3. At the highest powers, where the background saturation becomes significant, the line shape is hard to fit with a Lorentzian profile. However the Lorentzian fit at low and intermediate intensities is very good, showing the homogeneous nature of

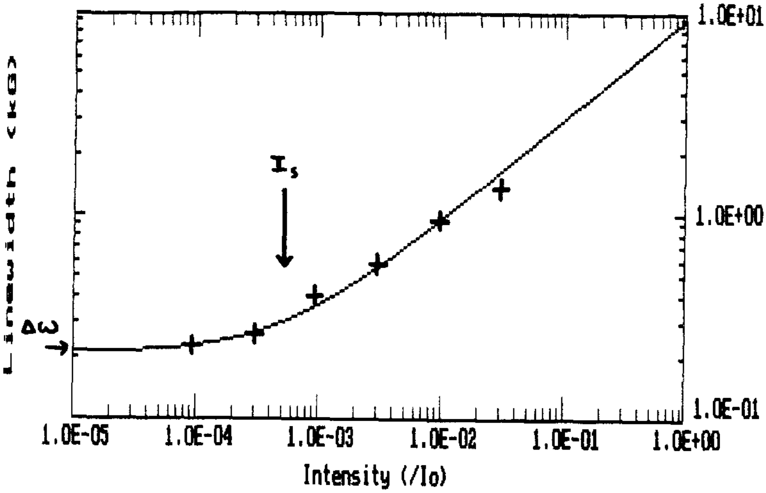

Figure 6. Linewidth of 1s-2p$^+$ transition as a function of intensity. Curve is a theoretical fit.

the transition. We plot, in figure 6, the linewidth, in field units, versus intensity in units of peak intensity. The fit curve is the standard homogeneous power broadening, of linewidth $\Delta\Omega$, for a two level system.

$$(\Delta\Omega)^2 = (\Delta\omega)^2 \ (1 + I/I_s)^{1/2} \qquad \text{Eqn. 9}$$

Our fit gives a low power linewidth, $\Delta\omega$, of 0.21 kG and a saturation intensity, I_s, of 1.0 x 10^{-5} I_o. The saturation intensity agrees reasonably well with our value determined from τ_2.

Turning away from the resonant behavior, complete magnetic field scans of the peak photoconductivity were taken as a function of incident power. Figure 7 shows some of the data at various magnetic fields. The data was fit using a two level model (equ 6) and normalized by the saturation intensity (I_s) and conductivity at infinite power levels, G_∞. As explained previously, the accuracy of the fit demonstates the applicability of our model to this case. Extending the magnetic field range from 0 to 4T, we may now plot our two fit parameters, $1/I_s$, G_∞, and G_∞/I_s, versus magnetic field as shown in figure 8. Note the position of the 1s-2p$^+$ transition and cyclotron resonance and hence the breakdown of our model in these regions. A

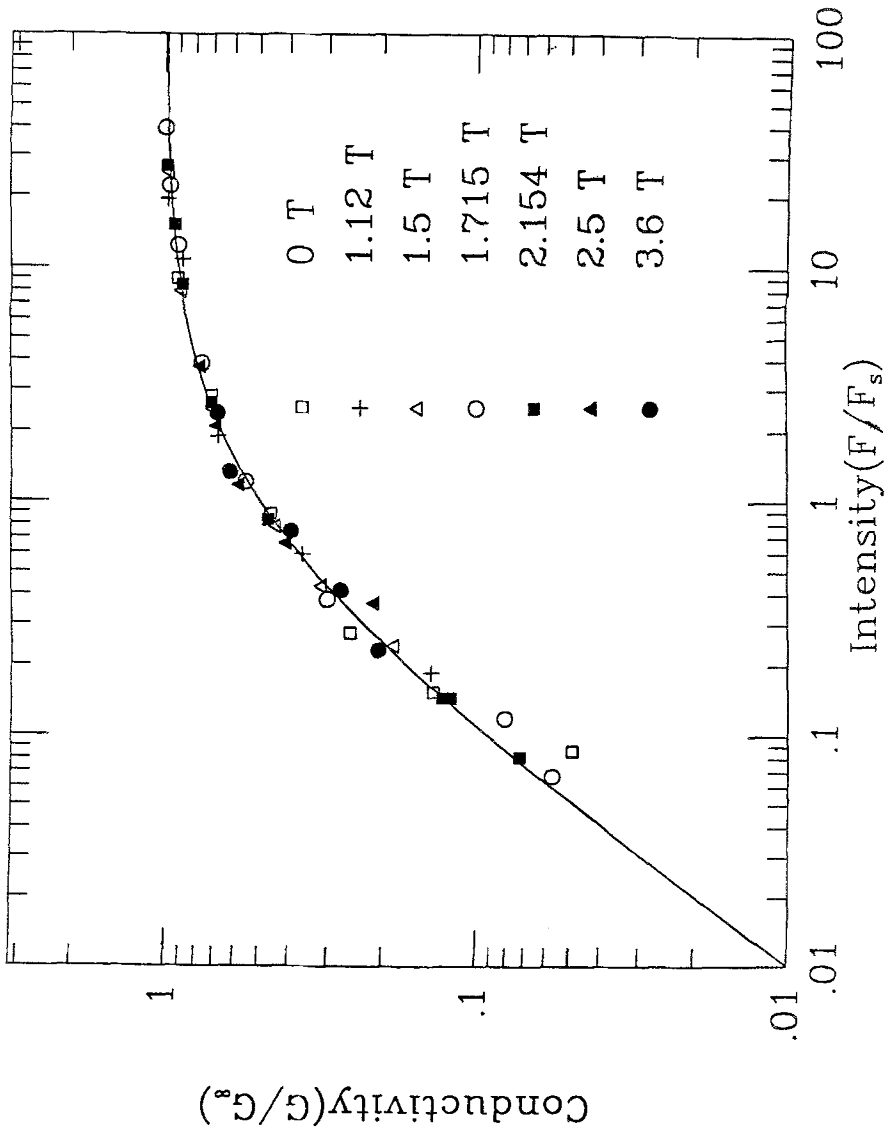

Figure 7. Photoconductance as a function of intensity at the indicated magnetic fields in units of peak conductance (G_∞) and saturation intensity (I_s). Curve is standard two level saturation behavior.

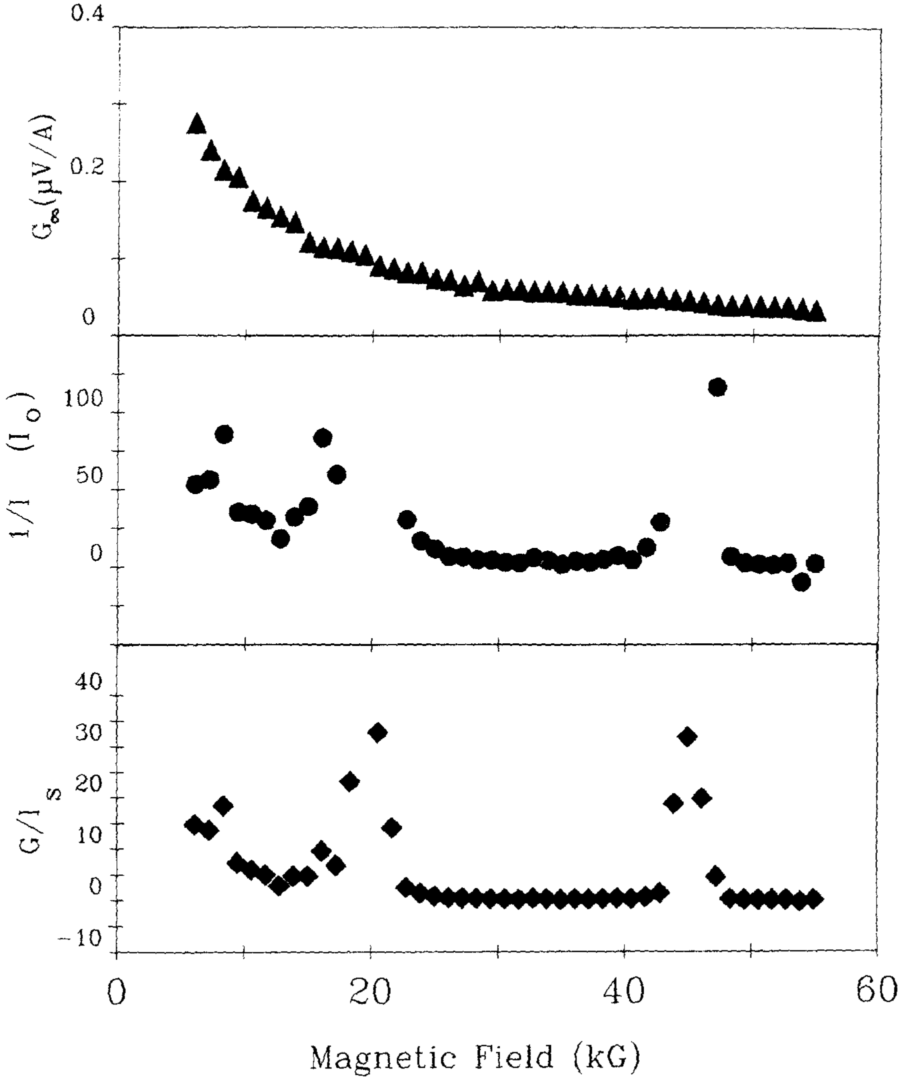

Figure 8. Fit parameters, $1/I_s$, G_∞, and G_∞/I_s, as a function of magnetic field using two level model. Note positions of $1s\text{-}2p^+$ and cyclotron resonance.

magnetic fields of particular interest should be the 1s-continuum edge, however for the given wavelength of 164 μm the Landau level edge is too close to the 1s-2p$^+$ resonance to be detectable. Nevertheless, the overall behavior seems to indicate a monotonically decreasing background saturation intensity and G_∞.

We may now make an alternate estimate of τ_1 and σ_c. Given $I_s = \hbar\omega(\sigma_c\tau_1)^{-1}$ and $G_\infty = e\mu P_a\tau_{(capture)}/\tau_1$, we can use the transient photoconductivity time to give an upper bound on $\tau_{(capture)} \leq 10$ nsec for all magnetic fields used. Furthermore, G_∞/I_s is directly proportional to σ_c and, using the constant value of $\tau_{(capture)} = 10$ nsec, we can find τ_1 and σ_c versus magnetic field. Note at B = 2.05 T, $\tau_1 = 117$ nsec and $\sigma_c = 8.6\text{x}10^{-14}$ cm^2, in reasonable agreement with our calculated values. Using faster switching times and detection electronics will allow us to map τ_1 and σ_c over all magnetic fields.

CONCLUSION

In summary, we have determined the relaxation lifetime and ionization probability of the 2p$^+$ donor state and the recombination lifetime of electrons in the N=0 Landau level. Power broadening was observed over 4 orders of magnitude in FIR intensity. Furthermore, the complete saturation behavior of the 1s-continuum transitions was characterized from 0 to 4T. These investigation show that much more detail of the electron kinetics may be obtained from saturation measurements using the UCSB FEL. Since the UCSB FEL is a high power tunable FIR source with sufficiently long pulse widths, non-linear spectroscopy of high purity semiconductors need no longer be limited to the few molecular lines available.

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REFERENCES

[1] T. Murotani and Y. Nishida, J. Phys. Soc. Jpn. 32, 986 (1972)

[2] E. Gornik, T.K. Chang, T.J. Bridges, V.T. Nguyen, I.D. McGee, and W. Muller, Phys. Rev. Lett. 40, 1151 (1978)

[3] K. Muro, N. Yutani, and Sh. Narita, J. Phys. Soc. Jpn. 49, 593 (1980)

[4] C.R. Pidgeon, A. Vass, G.R. Allan, W. Prettl, and L.A Eaves, Phys. Rev. Lett. 50, 1309 (1983)

[5] W. Prettl, A. Vass, G.R. Allan, and C.R. Pidgeon, Int. J. Infrared and Millimeter Waves 4, 561 (1983)

[6] G.R. Allan, A. Black, C.R. Pidgeon, E. Gornik, W. Seidenbusch, and P. Colter, Phys. Rev. B31, 3560 (1985)

[7] M. Weisspfenning, I. Hoeser, W. Bohm, W. Prettl, Phys. Rev. Lett. 55, 754 (1985)

[8] L. Elias, G. Ramian, J. Hu, and A. Amir, Phys. Rev. Lett., 57, 424 (1986)

[9] E. Scholl, W. Heisel, and W. Prettl, Z. Phys. B47, 287 (1982)

[10] C.J. Armistead, P. Knowles, S.P. Najda, and R.A. Stradling, J. Phys. C17, 6415 (1984)

[11] J. Simola and J. Virtamo, J. Phys. B11, 3309 (1978)

[12] W.S. Boyle and R.E. Howard, J. Phys. Chem. Solids 19, 181 (1961)

[13] J. Shah, Solid State Electron. 21, 43 (1978)

[14] K. Moser and W. Prettl, Int. J. Infrared and Millimeter Waves 7, 147 (1986)

[15] H. Hasegawa and R.E. Howard, J. Phys. Chem. Solids 21, 179 (1961)

[16] FIR from the FEL was switched off by irradiating a normally FIR transparent Si wafer with a high power Nd:YAG laser. The wafer is made totally reflecting in ≈10 nsec.

[17] F. Brown, A. Adamson, and P.A. Wolff, Int. J. Infrared and Millimeter Waves 1, 277 (1980)

[18] V.N. Zverev and D.V. Shovkun, Sov. Phys. JETP 60, 1003 (1984)

[19] J.M. Chamberlain, A.A. Reeder, L.M. Claesen, G.L.J.A. Rikken, and P. Wyder, Phys. Rev. B31, 2391 (1987)