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Free electron laser saturation spectroscopy of neutral donors and negative donor ions confined in GaAs/AlGaAs quantum wells

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Saturation of the D^0 1s-2p$^+$ transition, the D^--singlet transition and CR has been studied in donor (Si)-doped GaAs/AlGaAs multiple-quantum-well samples by magneto-transmission and magneto-photoconductivity measurements with the UCSB free electron laser. Effective lifetimes of the D^0 1s-2p$^+$transition were found to vary systematically with laser frequency, decreasing from 62 ns at 84 cm^{-1} to 3 ns at 124 cm^{-1}. The absorption coefficient of the D^--singlet transition initially increased by up to 40% and showed complete quenching at higher laser powers.

Key words: GaAs/Al$_x$Ga$_{1-x}$ quantum wells, donor impurities, saturation spectroscopy, free electron laser.

High-power far infrared (FIR) lasers enable the application of non-linear spectroscopic methods in the far infrared spectral range. Investigations of non-linear optical phenomena including the saturation of cyclotron resonance and impurity transitions in semiconductors have been carried out for many years. Saturation spectroscopy has been used to determine the lifetime of confinement states in multiple quantum well (MQW) structures [12], shallow hydrogenic impurity states in bulk semiconductors [1, 3, 4, 11], and the lifetime of Landau levels in bulk GaAs [1, 13] , InSb [14] and in GaAs/AlGaAs single heterostructures [2, 15] and very recently in GaAs/AlGaAs MQWs [16, 17]. Most of the early work on saturation was restricted to relatively low intensities for continuous wave (CW) or relatively short pulse widths for higher intensities. Only recently have tunable high power FIR light sources like free electron lasers (FEL) become available to researchers.

We have applied FEL saturation spectroscopy to doped quantum well (QW) to investigate lifetimes of various impurity states in semiconductor quantum wells. Two donor (Si)-doped GaAs/AlGaAs multiple-quantum-well samples (well width 200 Å) were investigated with the Free Electron Laser at the University of Santa Barbara (FEL at UCSB): A well-center-doped sample (Si at 2×10^{10} cm^{-2}), S1; and a well-and-barrier-doped sample (Si well/barrier $-2/3.5 \times 10^{10}$ cm^{-2}), S2. Both transmission and photoconductivity

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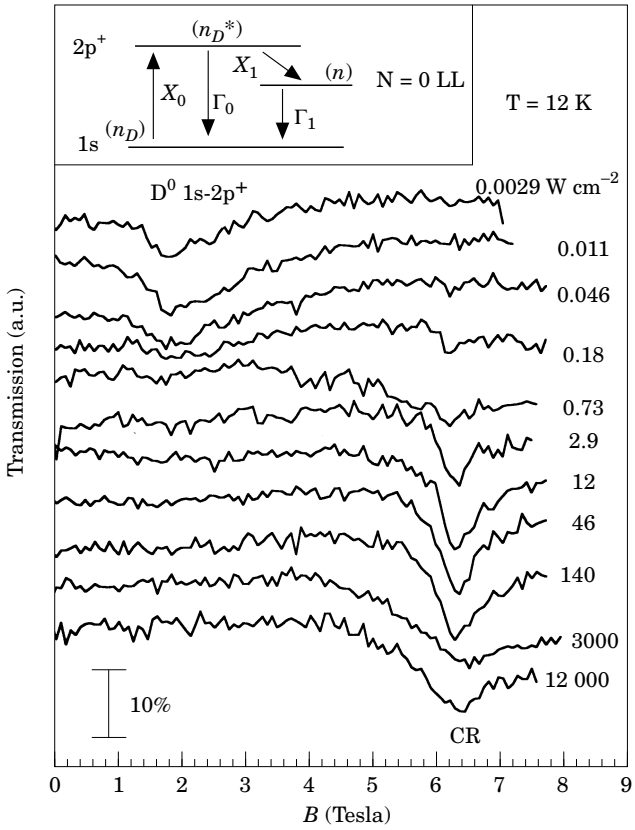

Fig. 1. Transmittance versus B for S1 at 84 cm^{-1} for several FEL laser power densities at 12 K. Saturation of the D^0 1s-2p$^+$ transition and CR is observable. Inset: Schematic of the three-level model discussed in the text.

measurements were performed simultaneously for the D^0 1s-2p$^+$ transition in S1. The long pulse width (2.6 μs) of the FEL, which is much longer than the expected lifetime (order of 10 ns) of the excited states of donors, assured a quasi-steady state condition of the system. The FEL is tunable and is capable of very high intensity (up to tens of kW cm^{-2}) over the wave length range from 1mm to 70 μm.

Transmission versus magnetic field data for S1 for various FEL power densities at 84 cm^{-1} are plotted in Fig. 1. At the lowest FEL power (2.86 mW cm^{-2}), only the D^0 1s-2p$^+$ transition is observable, as expected in this well-only-doped sample at low temperature (12 K). As the laser intensity is increased, the change in transmission of the D^0 1s-2p$^+$ transition decreases and there is no observable line above 12 W cm^{-2} ; CR appears at 183 mW cm^{-2} and increases in strength up to 137 W cm^{-2}, and then starts to saturate with apparent line-width broadening. This behavior is qualitatively the same as that of bulk donors in GaAs [1]. Similar behavior was observed at 114 cm^{-1} and 124 cm^{-1}. Due to the limited magnetic field (8 T), it was possible to study only the saturation of the D^0 1s-2p$^+$ transition for these laser lines.

We analyse the results with a three-level model in which an electron excited into the 2p$^+$ state (rate X_0) may either relax directly back to the 1s ground state (rate Γ_0), or may be transferred directly to the $N = 0$ Landau level (LL) (rate X_1 times the fraction of empty states $(g - n)/g$ in the $N = 0$ LL, where g and n are Landau level degeneracy and the density of electrons in the $N = 0$ LL, respectively) and then be captured by an ionized donor (rate Γ_1 times the fraction of ionized donors p_D/N_D). A schematic diagram of the model for $B > 4$ T is shown in the inset to Fig. 1. This model is identical to that used to analyse the bulk D^0 1s-2p$^+$

transition [1, 11], but the parameters should be different, since in 3D each Landau level has a continuum of states (corresponding to $k//B$) above the bottom, which lies at $E = (n + 1/2)\omega_c$, whereas in quasi-2D each Landau level is discrete with degeneracy g in the absence of broadening. The rate equations for this model may be written as

$$dn_D/dt = -X_0 n_D + X_0 n_D^* + \Gamma_0 n_D^* + \Gamma_1 n(p_D/N_D), \tag{1}$$

$$dn_D^*/dt = X_0 n_D - X_0 n_D^* - \Gamma_0 n_D^* - X_1 n_D^*((g-n)/g), \tag{2}$$

$$dn/dt = X_1 n_D^*((g-n)/g) - \Gamma_1 n(p_D/N_D), \tag{3}$$

where n, n_D, n_D^*, and p_D are the sheet densities of electrons in the $N = 0$ Landau level, the 1s ground state and $2p^+$ excited states of the neutral donor, and of ionized donors (D^+), respectively. The second term in eqns (1) and (2) represents stimulated photon emission, and we have ignored spontaneous photon emission from $2p^+$ to 1s states. The excitation rate is given by

$$X_0 = \sigma I/\hbar\omega, \tag{4}$$

where σ, I, and ω are the optical cross section, the intensity of the laser beam just inside the sample, and the photon energy, respectively. The absorption coefficient for the 1s-$2p^+$ transition in the presence of radiation may be written as

$$\alpha = \sigma(n_D - n_D^*), \tag{5}$$

and making use of eqns (1)–(5) in the steady-state, $dn/dt = dn_D^*/dt = dn_D/dt = 0$, with the condition of conservation of the total density of donors (N_D) and electrons (N_0), it can be expressed as a function of the laser intensity. It is instructive to consider some limiting cases. First, if $\Gamma_1 \gg X_1$, electrons in the $N = 0$ LL would relax with rate Γ_1 which is much faster than the electron transfer rate from the $2p^+$ state (X_1). In this case electrons do not accumulate in the $N = 0$ LL, and this system is effectively a two level system with two parallel relaxation processes with rates Γ_0 and X_1. This is the usual saturation process which is determined by the equilibrium between the pure absorption and the stimulated emission processes between the ground and the excited states. At sufficiently high laser intensity, half of the electrons in the ground state will be transferred to the excited state. The effective lifetime of the saturation process is, $\tau_{eff} \approx (\Gamma_0 + X_1)^{-1}$. The absorption coefficient is given by

$$\alpha = \alpha_0/(1 + I/I_s), \tag{6}$$

where the saturation intensity, $I_s = \hbar\omega/2\sigma\tau_{eff}$, is the laser intensity when the absorption coefficient becomes the half the linear value. These two processes are not experimentally distinguishable; therefore, the rates cannot be obtained separately from experimental data. In the other extreme case, $\Gamma_1 \ll X_1$, electrons in the $N = 0$ LL relax to the donor ground state (1s) with rate Γ_1 which is much slower than the electron transfer rate (X_1) from the $2p^+$ state. Therefore electrons accumulate in the $N = 0$ LL at high laser intensity. In this case, the saturation process is very different from the usual two level saturation; i.e. it is dominated by electron transfer from the ground state to the $N = 0$ LL, and the density of electrons in the excited state and the effect of stimulated emission are negligible. At sufficiently high laser intensity, *all* the electrons in the ground state are eventually transferred to the $N = 0$ LL in contrast to the usual case, for which only *half* of the electrons are transferred to the excited state. In the case of $\Gamma_1 \sim X_1$, the saturation process is a combination of these two limiting cases. At very high laser intensity, electrons are distributed among all three levels with the same order of magnitude; i.e. some of the electrons accumulate in the $N = 0$ LL with an equal density of electrons in the ground and excited states of donors. It should be noted that except for the first case, the saturation process cannot be described by an effective lifetime (τ_{eff}).

Results for the absorption coefficient versus laser power density are shown in Fig. 2 for three FEL photon energies. The peak absorption coefficient α_0 at low laser power density increases as the resonance field B increases (or as the energy of the laser line increases), as expected for the D^0 1s-$2p^+$ transition. Due to the

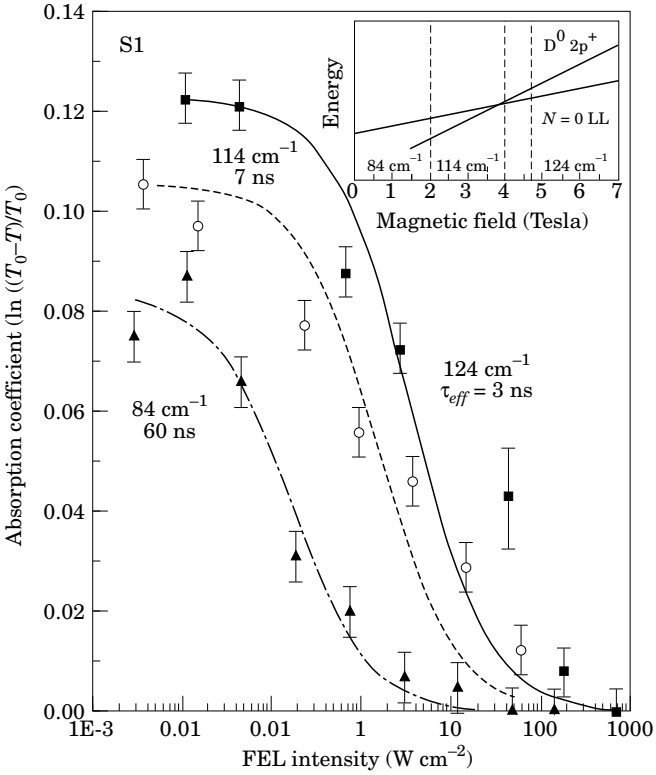

Fig. 2. Absorption coefficient of the D^0 1s-2p$^+$ transition for S1 as a function of FEL power density at various laser lines. The solid lines are the best fit from eqn (6). Inset: energy level diagram for the D^0 2p$^+$ state and the $N = 0$ LL for a 200 Å GaAs QW.

scatter of the absorption strength data at 114 cm^{-1} and 124 cm^{-1} with the FEL at low laser power, the values used for fitting were fixed by linear spectroscopy measurements with an optically pumped FIR laser and a BOMEM FTIR spectrometer. If we assume that the saturation process can be described by an effective lifetime, which is equivalent to the condition, $\Gamma_1 \gg X_1$, as explained above, the effective lifetime, τ_{eff}, of the D^0 1s-2p$^+$ state can be deduced from the data by fitting with eqn (6). The values of τ_{eff} obtained in this measurement decrease as laser frequency (and thus the magnetic field for resonance) increases (60 ns at 84 cm^{-1} to 3 ns at 124 cm^{-1}).

Now let us discuss the physical meaning of τ_{eff}. The direct 2p$^+$ to 1s lifetime via phonon emission for bulk GaAs is calculated to be of the order of 10 μs [18, 19], and it is assumed to have a similar order of magnitude for the quasi-2D case, much longer than the effective lifetimes measured here. If we assume $(\Gamma_1)^{-1} \ll (X_1)^{-1}$, the rate-limiting process is the transfer of electrons into the $N = 0$ LL, and $\tau_{eff} \approx (X_1)^{-1}$. In this case, the qualitative behavior of τ_{eff} can be understood by considering the energy separation between the D^0 2p$^+$ state and $N = 0$ LL as shown in the inset to Fig. 2. The $N = 0$ LL and the 2p$^+$ level cross near 4 T. Therefore the decay process from the 2p$^+$ to the $N = 0$ LL for $B < 4$ T is governed by absorption of acoustic phonons, and for $B > 4$ T by emission of acoustic phonons. At low temperature ($\sim$10 K) the number of acoustic phonons in the system is small, which means that the acoustic phonon emission process will dominate over the absorption process. Therefore relaxation from the 2p$^+$ state should be faster for fields > 4 T than for fields < 4 T. The observation of a very long lifetime at 84 cm^{-1} compared to that at 114 cm^{-1} and 124 cm^{-1} is probably due to the larger energy separation between the $N = 0$ LL and the D^0 2p$^+$ state. Our results for the saturation of

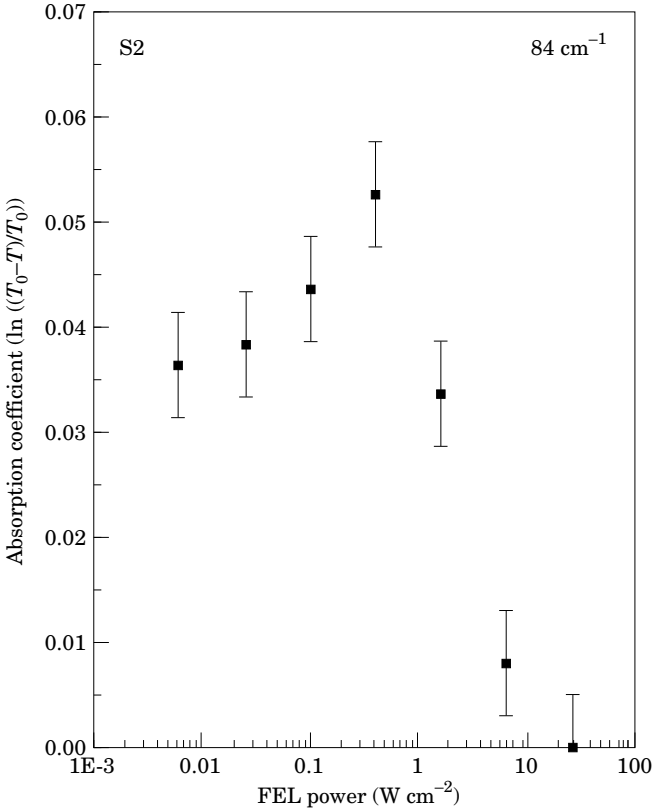

Fig. 3. Absorption coefficient of the D^- singlet transition for S2 at 84 cm^{-1} as a function of FEL intensity.

the D^0 1s-2p$^+$ transition in MQWs show similar behavior to that in uncompensated bulk GaAs [1], but they are significantly different from that in the compensated (high value of acceptor concentration, N_A, therefore large concentration of ionized donors, p_D, at low temperature) case [11].

For the 84 cm^{-1} line the data are fit reasonably well with a τ_{eff}. Different behavior is found for the 114 cm^{-1} and 124 cm^{-1} lines in Fig. 2. Eqn (6) does not give a very good overall fit to the data for these cases. The measured absorption coefficients are systematically lower than the fitted curve at low (below I_s) laser power, and systematically higher at high laser power. This can not be explained by simply adjusting I_s. It is clear from eqn (6) that for a given α_0, varying I_s does not change the functional form of α; i.e. it can only change the offset in the power density on the logarithmic scale, but it cannot change the overall width of the saturation range. To understand this behavior we have performed a full numerical optimization with the 3-level model. The numerical results show that the experimental data at 114 cm^{-1} and 124 cm^{-1} can be simulated reasonably only if a significant electron density accumulates in the $N = 0$ LL at high laser intensity (or equivalently $\Gamma_1 \leq X_1$). We believe that this process dominates at fields for which E(2p$^+$) $\geq$ E($N = 0$ LL). In this case, we can obtain an estimate for Γ_1: 3 ns $\leq (\Gamma_1)^{-1} \approx 10$ ns $\ll 60$ ns. Due to scatter in the data, the rates X_1 and Γ_0 are poorly defined.

Photoconductivity measurements of the D^0 1s-2p$^+$ transition were also carried out for S1 at the same time (*in situ*) as transmission, and show rather different behavior; i.e. even though the transmission data show complete quenching, the photoconductivity measurements still show significant transition strength at

high laser intensities. This difference is due to the fact that the photoconductivity signal is proportional to the density of electrons in the $N = 0$ LL, but is independent of the stimulated emission process, while transmission measurements probe *net* absorption; i.e. the net results of the difference between the pure absorption process and the stimulated emission process. Therefore, at high incident laser intensity, the two measurements give complementary information.

The laser intensity dependence of the transmission of the D^--singlet transition was studied in the barrier-and-well-doped sample, S2, at 84 cm^{-1} and saturation was observed. However, the absorption coefficient showed very different behavior than that of the D^0 1s-$2p^+$ transition; it initially increased by $\sim$ 40% and then showed complete quenching at higher laser intensities as shown in Fig. 3. The initial increase of the absorption coefficient for the D^- singlet transition is not due to activation of additional D^- sites. It may be due to deformation of the D^- wavefunctions by the strong AC electric field of the laser beam; however, this can only be considered to be speculation at present. The effective lifetime was estimated to be $\sim$ 3 ns from the saturation intensity. Detailed analysis of the saturation of the D^--singlet transition and the photoconductive detection of the saturation of the D^0 1s-$2p^+$ transition will be published elsewhere.

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References

[1] G. R. Allan, A. Black, C. R. Pidgeon, E. Gornik, W. Seidenbusch and P. Colter, Phys. Rev. **B 31**, 3560 (1985).
[2] M. Helm, E. Gornik, A. Black, G. R. Allan, C. R. Pidgeon, K. Mitchell and G. Weiman, Physica **134B**, 323 (1985).
[3] H. J. A. Bluyssen, J. C. Maan and P. Wyder, Solid State Comm. **31**, 465 (1979).
[4] T. Murotani and Y. Nisida, J. Phys. Soc. Japan **32**, 986 (1972).
[5] R. J. Sladek, J. Phys. Chem. Solid **5**, 157 (1958).
[6] H. Miyazawa and H. Ikoma, J. Phys. Soc. Japan **23**, 290 (1967).
[7] S. D. Ganichev, A. P. Dmitriev, S. A. Emel'yanov, Ya.V. Terent'ev, I. D. Yaroshetskii and I. N. Yassievich, Sov. Phys. JETP **63**, 256 (1986).
[8] R. L. Green and K. K. Bajaj, Phys. Rev. **B 31**, 913 (1985).
[9] A. B. Dzyubenko and A. Y. Sivachenko, Phys. Rev. **B 48**, 14690 (1993).
[10] J.-P. Cheng, Y. J. Wang, B. D. McCombe and W. Schaff, Phys. Rev. Lett. **70**, 489 (1993).
[11] C. R. Pidgeon, A. Vass, G. R. Allan, W. Prettl and L. Eaves, Phys. Rev. Lett. **50**, 1309 (1983).
[12] F. H. Julien, J-M. Lourtioz, N. Herschkorn, D. Delacourt, J. P. Pocholle, M. Papuchon, R. Planel and G. Le Roux, Appl. Phys. Lett. **53**, 116 (1988).
[13] K. Mitchell, A. Black, C. R. Pidgeon, G. R. Allen, M. F. Kimmit and E. Gornik, J. Phys. C: Solid State Phys. **20**, 5217 (1987).
[14] E. Gornik, T. Y. Chang, T. J. Bridges, V. T. Nguyen, J. D. McGee and W. Muller, Phys. Rev. Lett. **40**, 1151 (1978).
[15] G. A. Rodriguez, R. M. Hart, A. J. Sievers, F. Keilmann, Z. Schlesinger, S. L. Wright and W. I. wang, Appl. Phys. Lett. **49**, 458 (1986).
[16] R. Ranganathan, J. Kaminski, W. J. Li, J-P. Cheng and B. D. McCombe, Proc. SPIE **1678**, 120(1992).
[17] W. J. Li, B. D. McCombe, J. P. Kaminski, S. J. Allen, M. I. Stockman, L. S. Muratov, L. N. Pandey, T. F. George and W. J. Schaff, Semicond. Sci. Technol. **9**, 630 (1994).
[18] K. Muro, N. Yutani, and S. Narita, J. Phys. Soc. Jpn. **49**, 593 (1980).
[19] G. Ascarelli and S. Rodriguez, Phys. Rev. **C 12**, 2809 (1979).