Nonlinear Far-Infrared Magnetoabsorption and Optically Detected Magnetoimpurity Effect in *n*-GaAs

C. R. Pidgeon, A. Vass, and G. R. Allan
Department of Physics, Heriot-Watt University, Edinburgh EH9 3JZ, United Kingdom

and

W. Prettl
Department of Physics, University of Regensburg, D-8400 Regensburg, West Germany

and

L. Eaves
Department of Physics, University of Nottingham, Nottingham NG7 2RD, United Kingdom
(Received 18 January 1983)

Measurements have been made of cw nonlinear far-infrared magnetoabsorption in pure, partially compensated n-GaAs. A new optical observation of the magnetoimpurity resonance has been demonstrated. Analysis in terms of a three-level model gives effective lifetimes of $\sim 0.5\ \mu$s "off resonance" and 30 ns "on resonance," implying the possibility of laser action in the former case.

PACS numbers: 78.50.Ge, 42.65.Gv, 78.20.Ls

We report the first measurement of cw far-infrared (FIR) nonlinear magnetoabsorption, and optically detected magnetoimpurity resonance (OMIR) in epitaxial n-GaAs. Up to the present time, apart from one experiment in n-InSb using a cw HCN laser,[1] FIR nonlinear magnetoabsorption has been confined to a few measurements with CO_2-laser-pumped systems.[2,3] While this may have provided an accurate probe of saturation effects in n-InSb, we show that this is not the case for n-GaAs where the measured state lifetimes are comparable with, or even longer than, the CO_2-laser pulse. We have determined effective lifetimes for shallow $2p \to 1s$ impurity transitions which differ according to whether the OMIR is "on resonance" or "off resonance." In the former case τ_{eff} is of ~50 ns; in the latter τ_{eff} is of ~0.5 μs. The unusually long decay time in the off-resonance case is in accord with the suggestion that laser action may take place between the donor impurities in n-GaAs.[4] We would like to stress that the observation of a marked dependence of the saturation behavior on magnetic field is of fundamental importance for the subject of the optical kinetics of extrinsic impurity states in semiconductors.

We have used a high-intensity Edinburgh Instruments optically pumped far-infrared laser—operated either cw or with controlled pulse lengths —to measure impurity magnetoabsorption in epitaxial n-GaAs. Results showing strong saturation of the $1s$-$2p_+$ transition as a function of laser intensity at three different wavelengths are illustrated in Fig. 1, for partially compensated material ($N_D - N_A = 4.1 \times 10^{14}$ cm^{-3}, thickness 40 μm). The general effect is seen for powers some ten times lower than in an earlier CO_2-laser experiment.[3] An additional feature is the dramatic modulation with wavelength and magnetic field of the saturation intensity from a high (at 186 μm, 14 kG) through low (at 164 μm, 20 kG) to high value (at 152 μm, 24 kG), discussed in detail below. We regard this as a direct optical manifestation of the magnetoimpurity resonance seen previously only in transport measurements.[5]

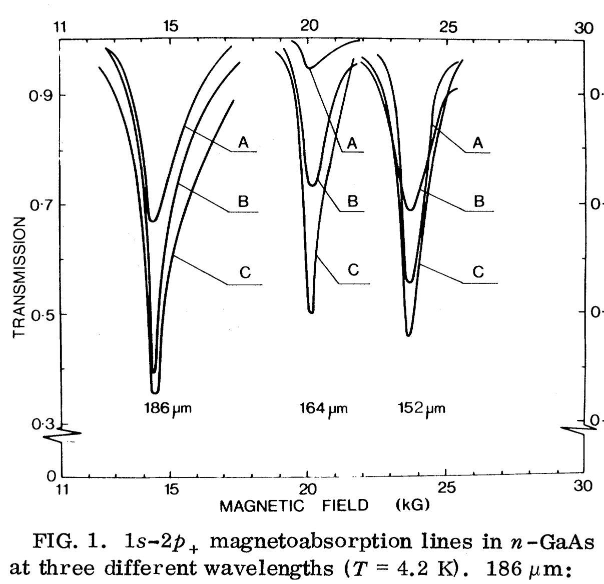

FIG. 1. $1s$-$2p_+$ magnetoabsorption lines in n-GaAs at three different wavelengths (T = 4.2 K). 186 μm: A = 33, B = 10, C = 1.8 mW cm^{-2}; 164 μm: A = 27.6, B = 4.3, C = 0.1 mW cm^{-2}; 152 μm: A = 16.6, B = 8.1, C = 1.6 mW cm^{-2}.

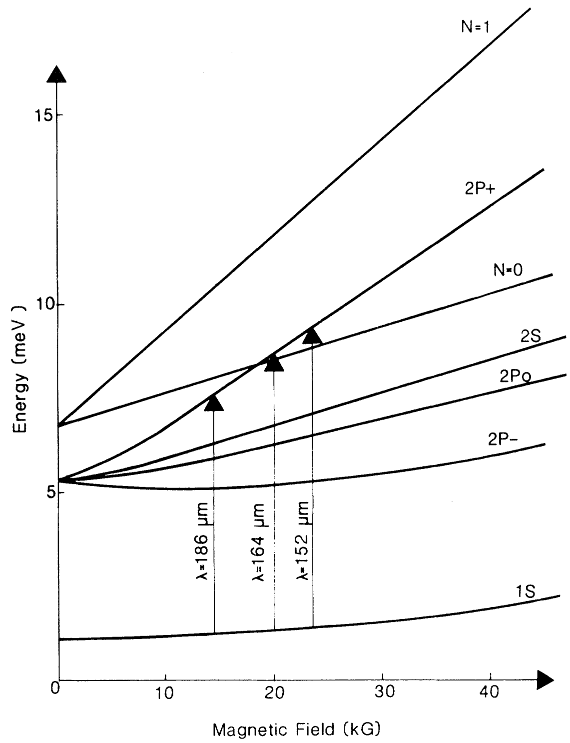

FIG. 2. Plot of magnetic field dependence of shallow impurity states and Landau levels for n-GaAs (after Ref. 6). The $1s$-$2p_+$ transition, measured at three energies is also shown.

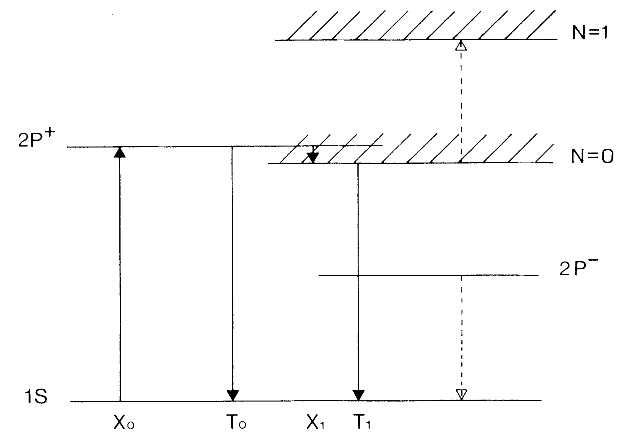

FIG. 3. Schematic diagram of $1s$, $2p_+$, and $N=0$ Landau subband three-level system for magnetic field 24 kG. Transition rates are described in the text. $2p_-$ and $N=1$ levels responsible for the OMIR are also shown.

The $1s$-$2p_+$ transition, observed at the three laser wavelengths, is shown together with the computed impurity and conduction-band Landau levels in Fig. 2.

We interpret our results for an n-type, partially compensated semiconductor, with a three-level model,[1,2] where an electron excited into the $2p_+$ state (rate X_0) may either relax directly into the $1s$ ground state (rate T_0), or be transferred directly to the conduction band (rate X_1) and then be captured by an ionized donor (rate T_1 times the concentration of ionized donors). The situation is shown schematically in Fig. 3 for the case at 24 kG (152 μm), where the $2p_+$ state is at a higher energy than the $N=0$ Landau level. Also shown are the $2p_-$ and $N=1$ Landau levels involved in the OMIR—see below. The rate equations are given by

$$dn/dt = X_1 n_D{}^* - T_1 n p_D\,, \tag{1}$$

$$dn_D{}^*/dt = X_0 n_D - T_0 n_D{}^* - X_1 n_D{}^*, \tag{2}$$

$$dn_D/dt = -X_0 n_D + T_0 n_D{}^* + T_1 n p_D, \tag{3}$$

where n, n_D, $n_D{}^*$, and p_D are the concentrations of electrons in the $N=0$ Landau subband, $1s$ ground state, and $2p_+$ excited state, and of ionized donors, respectively. At low temperatures the excitation rate is given by $X_0 = \sigma I/\hbar\omega$, where σ, I, and $\hbar\omega$ are the optical cross section, radiation intensity, and photon energy. The relaxation constant may be written $T_0 = \tilde{T}_0 + \sigma I/\hbar\omega$, where the first term refers to phonon emission and the second to stimulated photon emission (we ignore spontaneous emission). Depending whether the $2p_+$ state is below or above the $N=0$ Landau subband, the transfer rate X_1 of electrons to the band is governed by phonon absorption or emission, or by tunneling. The band to ground-state recombination is through s states, given by $T_1 = \langle v\rangle\sigma_c$, where $\langle v\rangle$ is the average velocity of electrons and σ_c the capture cross section of ionized donors.[7-9]

Under steady-state conditions, taking into account the conservation of total donor number, $N_D = n_D + n_D{}^* + p_D$, and charge, $P_A = N_D - N_A = n_D + n_D{}^* + n$, the carrier concentrations are given by

$$n_D = \frac{(T_0 + X_1)(P_A - n)}{T_0 + X_0 + X_1}\,, \tag{4}$$

$$n_D{}^* = \frac{X_0(P_A - n)}{T_0 + X_0 + X_1}\,, \tag{5}$$

$$T_1 n(N_A + n) = \frac{X_0 X_1 (P_A - n)}{T_0 + X_0 + X_1}\,. \tag{6}$$

In the linear limit, where $n < N_A$, this gives for the ($1s$-$2p_+$) absorption coefficient

$$\alpha = \sigma(n_D - n_D{}^*) = \alpha_0/(1 + I/I_s), \tag{7}$$

where

$$I_s = \frac{\hbar\omega}{2\sigma\tau_{\rm eff}} = \frac{\hbar\omega}{2\sigma}\,\frac{\tilde{T}_0 + X_1}{1+(X_1/2T_1N_A)}. \tag{8}$$

In Fig. 4 are shown plots of the intensity dependence of the peak absorption coefficient for measurements at 152 μm (24 kG), 164 μm (20 kG), and 186 μm (14 kG). The solid curves are the best fits from Eq. (6), yielding values of $\tau_{\rm eff}$ of 51.5 ns ($I_s = 22.1$ mW/cm^2), 228 ns ($I_s = 8.1$ mW/cm^2), and 32.5 ns ($I_s = 40$ mW/cm^2), respectively. Good fits are obtained except at the highest intensities where the linear limit of Eq. (6) probably breaks down.

Unfortunately, as pointed out by Muro, Yutani, and Narita,[3] the presence of long-lived states renders the steady-state anlysis invalid for the CO_2-laser-pumped FIR laser experiments of these authors on n-GaAs. Thus, in the absence of detailed information about line shapes it is not possible to deduce any information from their "apparent" saturation measurements. Furthermore, the samples were too thin to obtain absorption measurements so that results had to be deduced from photoconductivity measurements. It is even less clear how to interpret non-steady-state intensity-dependent measurements in this case. Finally, the two-level model used is certainly inadequate to explain results which include significant transfer of free electrons into and between conduction-band Landau levels in the conduction process.

The magnetoimpurity resonance (MIR) occurs in the longitudinal magnetoresistance of n-GaAs,

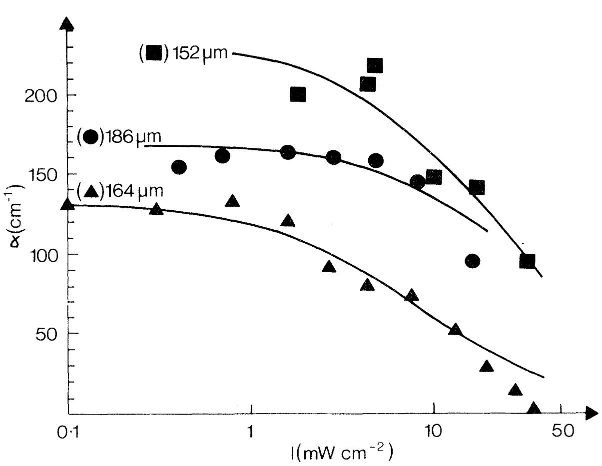

FIG. 4. Intensity dependence of $1s$-$2p_+$ magnetoabsorption in n-GaAs at three different wavelengths. Solid lines show the best fits from Eq. (6).

under hot-electron conditions, due to a scattering process in which resonant heating of conduction electrons from the $N=0$ to $N=1$ Landau levels is caused by the deexcitation of donors from the $2p_-$ to $1s$ ground states.[5] Hot-donor-electron conditions are required which can be produced either by white or laser light, of energy greater than the $2p_-$ to $1s$ energy (in our case, the FIR laser). The first and second resonances of the MIR series, given by $n\hbar\omega_c = \Delta E(1s\text{-}2p_-)$, occur at fields 24 and 12 kG, close to the fields of the more weakly saturating wavelengths (24 and 14 kG). We call this the OMIR effect, where the transitions causing resonant heating of the free carriers are shown by dashed lines in Fig. 3. The strongly saturating case (at 20 kG) is close to the middle of the off-resonance field (18 kG).

In interpreting the meaning of $\tau_{\rm eff}$ in Eq. (8) we can look to other work on n-GaAs, particularly in the off-resonant case. The conduction-band to $1s$ ground-state lifetime is dependent on the degree of compensation through N_A, and may vary from a few nanoseconds to several microseconds[4,8,10]; it is determined principally by the lifetime for the $2s$-to-$1s$ phonon transition. The $2p_+$-to-$1s$ lifetime is calculated to be of order 10 μs.[3,9] Thus $\tau_{\rm eff} \ll \tau_{2p_+,1s}$, and we measure $\tau_{\rm eff} \leq (X_1)^{-1} + (2T_1N_A)^{-1}$. That is, the saturation of intensities we determine gives the lifetime for the transfer of electrons from the $2p_+$ state via the conduction band to the $1s$ ground state, and hence an upper limit on τ_c. In purer material, where it has not been possible to measure magnetoabsorption in epitaxial films $[N_D - N_A \approx (1-4)\times 10^{13}$ cm$^{-3}]$, we have made photoconductivity measurements that show $\tau_{\rm eff}$ as long as a few microseconds.

In the resonant case electrons make transitions from the $N=0$ Landau subband to higher subbands ($N=1$, 2, etc.) through inelastic scattering processes in which the donor deexcites from the $2p_-$ state into the $1s$ ground state, thus reducing the saturation—i.e., raising T_1 and shortening $\tau_{\rm eff}$. The observation of this effect makes it clear that a substantial hot-electron population is built up in the long-lived $2p_-$ state, with very possibly a population inversion in the off-resonance condition. Under these circumstances, with large measured values of $\tau_{\rm eff}$, $2p_+$-to-$1s$ photon transitions can dominate $2s$-to-$1s$ phonon transitions, so that laser emission is achievable.

In conclusion, we have measured cw nonlinear FIR magnetoabsorption and OMIR in partially compensated n-GaAs, demonstrating long lifetimes of shallow impurity states, and the possibility of laser action between them.

We are very grateful to P. Colter of the U. S. Air Force Wright Aeronautical Laboratories for provision of thick epitaxial layers of n-GaAs, and to R. L. Aggarwal, E. Gornik, and R. A. Stradling for helpful discussion and suggestions. Two of us (A.V. and G.R.A.) have benefitted from Science and Engineering Research Council (U.K.) studentships. This work was supported in part by the Deutsche Forschungsgemeinschaft.

[1]T. Murotani and Y. Nisida, J. Phys. Soc. Jpn. 12, 986 (1972).

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

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

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

[5]L. Eaves and J. C. Portal, J. Phys. C 12, 2809 (1979).

[6]D. M. Larsen, J. Phys. Chem. Solids 29, 271 (1968).

[7]J. McManus, R. People, R. L. Aggarwal, and P. A. Wolff, J. Appl. Phys. 52, 4748 (1981).

[8]G. E. Stillman, C. M. Wolfe, and D. M. Korn, in *Proceedings of the Eleventh International Conference on the Physics of Semiconductors, Warsaw, 1972*, ited by The Polish Academy of Sciences (Elsevier, Amsterdam, 1972), p. 863.

[9]G. Ascarelli and S. Rodriguez, Phys. Rev. 124, 1321 (1961).

[10]H. J. A. Bluyssen, J. C. Maan, and P. Wyder, Solid State Commun. 31, 465 (1979).