PROC. 15TH INT. CONF. PHYSICS OF SEMICONDUCTORS, KYOTO, 1980 J. PHYS. SOC. JAPAN 49 (1980) SUPPL. A p. 593–596

> SATURATION OF ABSORPTION AND PHOTOCONDUCTIVITY DUE TO SHALLOW IMPURITIES IN SEMICONDUCTORS BY HIGH POWER FAR-INFRARED LASER BEAM

Kiyofumi Muro, Naoki Yutani and Shin-ichiro Narita

Department of Material Physics Faculty of Engineering Science Osaka University Toyonaka, Osaka 560, Japan

The saturation effect of far-infrared donor absorption in Ge has been observed by using the high power far-infrared laser. The lifetimes of the excited states of arsenic and antimony impurities are estimated to be 11 and 15*n*sec., respectively. The lifetime for the donor in GaAs is also estimated to be 20*n*sec. from the saturation of far-infrared photoconductivity. These results are discussed based on the theoretical investigations by Ascarelli and Rodriguetz, and also by Beleznay and Pataki.

A recent development of high power far-infrared (FIR) laser enables us to study various kinds of FIR non-linear optical phenomena: multiphoton absorption, stimulated Raman emission [1] due to impurities in semiconductors, four photon mixing due to free carriers in narrow-gap semiconductors, etc. Some experiments on the saturation of the cyclotron resonance in InSb have been already performed, and the relaxation mechanism of the Landau electrons has been discussed [2,3]. In the present work, we observe the saturation of FIR absorption and photoconductivity due to shallow impurities in order to investigate impurity states and their relaxation process in the presence of high power FIR laser radiation.

The high power FIR laser pumped by TEA CO<sub>2</sub> laser was employed as a strong FIR source. The laser was operated at 0.5Hz with a pulse width of 200n sec. The strongest laser lines; 90.6 and  $151.8\mu\text{m}$  of NH<sub>3</sub> and  $66\mu\text{m}$  of D<sub>2</sub>O, were mainly used. The experimental set-up is schematically shown in Fig.1. The FIR beam power was measured with a calibrated pyroelectric detector operating in a joule meter. Teflon plates with 3mm thickness or black polyethylene sheets were used as a beam attenuator, after calibration. The sample was mounted in the center of a super-conducting solenoid immersed in liquid He. The photoconductivity and transmission signals were recorded against the



Fig.l Experimental arrangement

magnetic field by the aid of the combination of a fast response bias circuit and a boxcar integrator.

The FIR magneto-absorption due to shallow donors in Ge was observed at liquid He temperature, changing the FIR beam intensity. The FIR spectra observed at 90.6 $\mu$ m and 4.2K in a magnetic field along [111]-crystal axis for a Ge sample with As donors of 1x10<sup>14</sup> cm<sup>-3</sup> are shown in Fig.2. The absorption due to the As impurity decreases with increasing





FIR beam intensity. The Zeeman absorption spectra of As and Sb donors in Ge were reported by Horii and Nisida [4,5]. According to their investigation, the



Fig.3 Saturation behaviours of absorption coefficients at 90.6µm for three absorptions in As-doped Ge

absorption peaks in Fig.2 are assigned to the transitions to  $3p_{+1}(A)$ ,  $2p_{+1}(A)$  and  $2p_{+1}(B)$ , respectively. The radiation beam intensity dependence of the absorption coefficients corresponding to three different excitations of As impurity is shown in Fig.3 for two Ge samples with the impurity concentrations of  $9 \times 10^{14}$  and  $1 \times 10^{14}$  cm<sup>-3</sup>. Similar measurements were done for a Ge sample containing Sb donors of  $1.4 \times 10^{14}$  cm<sup>-3</sup> by using 90.6 and 151.8 µm laser lines.

Reflecting the fact that the saturation behaviour of the absorption is almost independent of the donor density (see Fig.3), the electrons must be accumulated in excited states of donor under optical excitation. Then, the rate equation can be expressed as

$$\frac{\mathrm{d}\mathbf{n}_{\mathrm{d}}}{\mathrm{d}\mathbf{t}} = -W\mathbf{n}_{\mathrm{d}} + (N_{\mathrm{d}} - n_{\mathrm{d}})/\mathrm{T}, \tag{1}$$

where  $N_d$  is the donor density and  $n_d$ ; the ground state donor density during excitation. W is the optical transition probability expressed as  $W=\alpha_0 I/\hbar\omega N_d$  (I: the radiation beam intensity,  $\alpha_0$ : the absorption coefficient for a weak radiation beam), and T; the effective lifetime of the excited states into ground state.

Using the steady state condition, we obtain a simple equation describing the absorption coefficient for a strong radiation beam:

$$\alpha = \alpha_0 \left( \frac{1}{1 + WT} \right) = \alpha_0 \left( \frac{1}{1 + I/I_s} \right) , \qquad (2)$$

where  $I_s$  is the saturation beam intensity defined by  $\alpha = \alpha_0/2$ . The effective lifetime is estimated by the following equation:

$$T = \hbar\omega N_d / \alpha_0 I_s$$
 (3)

The saturation beam intensity and lifetime obtained for As and Sb

donors under some optical excitations are listed in Table I. The data represent no appreciable temperature dependence below 4.2K.

The relaxation of an electron between the excited states of donor was theoretically investigated by Ascarelli and Rodriguetz [8,9], and later Beleznay and Pataki [10] in order to describe the recombination cross-section of an electron and an ionized donor impurity in germanium [6,7]. According to their theory, the relaxation is governed by the interaction with a longitudinal acoustic phonon due to deformation potential and, the transition probability is larger for the transition between higher excited states than for the transition into ground state. Among the transitions into the ground state, the transition from 2s state is dominant.

The lifetimes, llnsec. for As donor and 15nsec. for Sb donor, obtained at zero magnetic field, are close to the inverse of the transition probability from 2s to 1s state, 8.5nsec., calculated by Beleznay and Pataki [10]. The effective lifetime of an excited electron into the ground state seems to be governed by the transition from 2s excited state to 1s ground state; as expected from the preceding theoretical results.

The lifetime for As donor obtained at 32kG under the excitation to  $2p_{+1}(B)$  is fairly smaller than the value at zero field. This result might be ascribed to the shrinkage of donor wavefunction in a magnetic field, for the phonon emitting probability depends strongly on the effective Bohr radius [8]. On the other hand, the lifetime (92*n*sec.) for Sb donor, obtained at 5kG by 151.8µm radiation for the excitation to  $2p_{-1}(A)$  is much larger than the other values, and is rather close to the theoretically estimated lifetime of 2p state into 1s state; 120*n*sec. [10]. This result seems to be associated with the theoretical prediction that the phonon relaxation is suppressed for low lying non-zero angular momentum state [8,10]. However, the presence of the magnetic field and the mass anisotropy in conduction band makes the problem very complicated, and we have no quantitative discussion. Further the value (92*n*sec.) is unreliable because the assumption of steady state condition becomes invalid for such a large value.

The photoconductivity in n-Ge, n-GaAs and n-InSb was also measured as a function of radiation beam intensity. The saturation behaviours of the photoconductivity and the absorption in Ge sample resemble each other and therefore the saturation of the photoconductivity is also considered to arise from the depletion of the electrons in the ground state. The radiation beam intensity dependence of the photoconductivity spectra of n-GaAs with residual donors of  $5 \times 10^{13}$  cm<sup>-3</sup> observed at 4.2K and  $151.8 \mu m$  is shown in Fig.4. The photoconductivity at 0 and 24kG corresponding to the excitations;  $ls \rightarrow c.b.$  and  $ls \rightarrow 2p_{+1}$ , respectively [12], is plotted against the radiation beam intensity as shown in the inset. The saturation beam intensity is estimated to be 1.8 and  $0.35W/cm^2$ , respectively.

excitation from ls to	Ge(As) 1x10 <sup>14</sup> cm <sup>-3</sup>		excitation	Ge(Sb) 1.4x10 <sup>14</sup> cm <sup>-3</sup>	
	$I_{s}(W/cm^{2})$	T(nsec.)	from ls to	$I_{s}(W/cm^{2})$	T(nsec.)
near c.b. at OkG	25	11	c.b. at OkG	20	15
2p <sub>+1</sub> (A) at 12kG	12	10	2p_1(A) at 5kG	0.67	92
2p <sub>+1</sub> (B) at 32kG	10	6			

Table. I. Saturation beam intensity  ${\rm I}_{\rm S}$  and effective lifetime T for arsenic and antimony donors in germanium



Typical photoconductivity Fig.4 spectra of high purity GaAs with residual donors of 5×1013 cm<sup>-3</sup> observed at 4.2K for three intensities of 151.8µm radiation: In the inset, the saturation behaviours of photoconductivities observed at 0 and 24kG are shown

The high purity GaAs epitaxial layer is too thin ( $\circ$ 10 $\mu$ m) to observe the absorption of donors, so we have no experimental value about the absorption coefficient. Therefore, we estimate the absorption cross-section  $(\alpha_0/N_d)$  of a donor in GaAs in the following methods. The absorption cross-section for  $ls \rightarrow 2p_{+1}$  transition at 24kG is estimated to be 9.3×10<sup>-13</sup> cm<sup>2</sup> for left circular polarized light according to eq.(17) in ref.[11], where the electric dipole matrix element is calculated by using the harmonic oscillator type wavefunctions for 1s and 2p+1 states with the variational parameters given by Narita and Miyao [12,13], and the spectral width obtained in the present experiment. The cross-section for 151.8 $\mu$ m at zero field is estimated to be 3.5 $\times$ 10<sup>-14</sup>cm<sup>2</sup> with the correspondence relation in the effective mass theory by utilizing the corresponding absorption cross-section (at 90µm) for a Sb donor in Ge.

Using the above values for the absorption cross-section  $(\alpha_0/N_d)$  the effective lifetime of excited electrons at 0 and 24kG is estimated to be 20 and 8.2nsec., respectively, according to eq.(3), taking account of the linear polarization of the laser beam. The obtained lifetime, 20nsec. at

zero field is about 1/400 of the valúe for the lifetime of 2s state, estimated from Ascarelli and Rodriguetz's theory. Therefore, we can presume that the relaxation of donor excited states in GaAs is governed rather by the piezo-electirc electron-phonon interaction than by the interaction due to the deformation potential treated in the preceding theory.

The saturation behaviour of the photoconductivity spectra in high purity n-InSb is quite similar to that of n-GaAs shown in Fig.4. The saturation beam intensity and lifetime obtained in the present photoconductivity measurement are in good agreement with the results referring to the impurity cyclotron resonance absorption [3].

## References

- 1) C.K.N.Patel and Y.Yafet: Appl. Phys. Letters 34 (1979) 318.
- 2) T.Murotani and Y.Nisida: J. Phys. Soc. Japan 32 (1972) 986.
- 3) E.Gornik, T.Y.Chang, T.J.Bridges, V.T.Nguyen and J.D.Mcgee: Phys. Rev. Letters 40 (1978) 1151.
- 4) K.Horii and Y.Nisida: J. Phys. Soc. Japan 31 (1971) 783.
- Y.Nisida and K.Horii: J. Phys. Soc. Japan 31 (1971) 776.
   S.H.Koenig: Phys. Rev. Letters 110 (1959) 988.
- 7) G.Ascarelli and S.C.Brown: Phys. Rev. 120 (1960) 1615.
- 8) G.Ascarelli and S.Rodriguetz: Phys. Rev. 124 (1961) 1321.
  9) G.Ascarelli and S.Rodriguetz: Phys. Rev. 127 (1962) 167.
- F.Beleznay and G.Pataki: Phys. Stat. Sol. 13 (1966) 499.
   R.F.Wallis and H.J.Boulden: J. Phys. Chem. Solids 7 (1958) 78.
- 12) S.Narita and M.Miyao: Solid State Commun. 9 (1971) 2161.
- 13) M.Miyao: his doctor thesis at Osaka University (1974).