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July 1986

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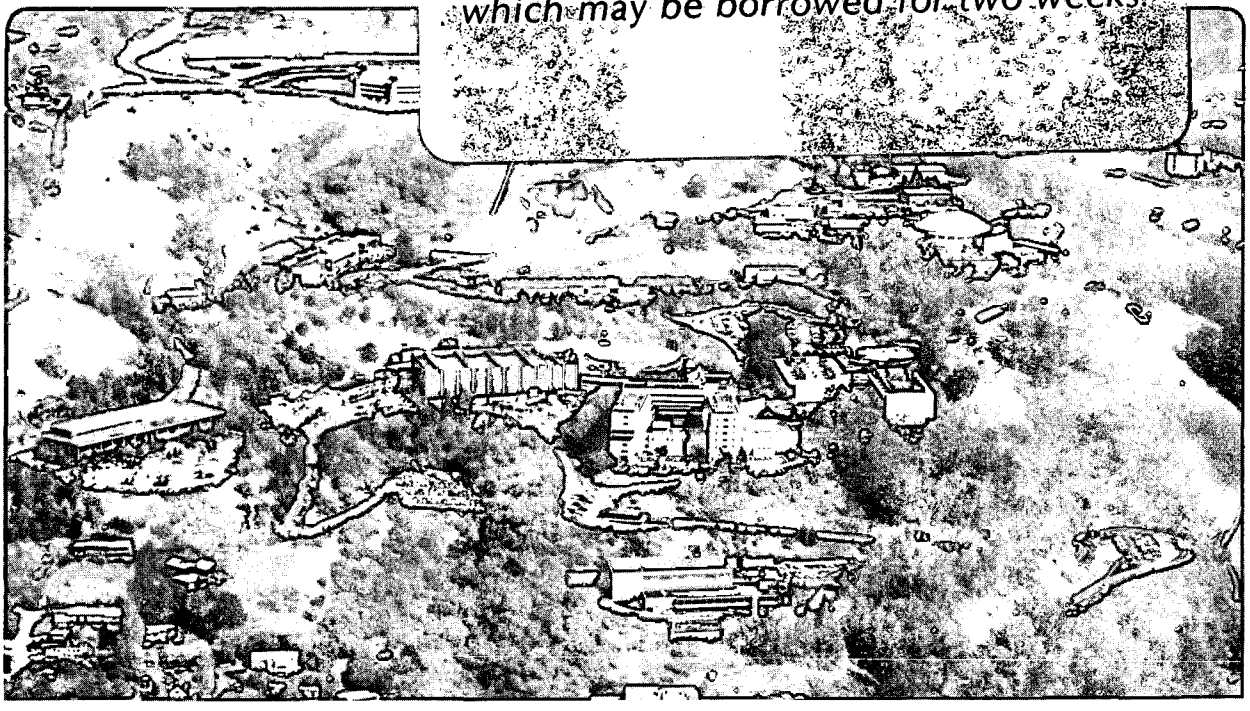

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Intrinsic Linewiths of 1S→nP Donor Transitions in Ultra-pure Germanium

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Intrinsic Linewidths of 1S→nP Donor Transitions in Ultra-pure Germanium

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Photon induced dipole transitions of electrons or holes bound to donor or acceptors respectively, belong to the most fundamental optical processes in a solid. The absorption lines produced by such transitions have fullwidths at half maximum (FWHM) which typically do not reflect intrinsic properties of the isolated impurity in the crystal. Random electric fields produced by ionized impurities, strain fields due to dislocations and other defects, overlap of the very extended hydrogenic bound states and the crystal temperature all contribute to the FWHM values which are significatively larger than theory predicts for the isolated impurity embedded in a perfect crystal[1,2].

In an attempt to experimentally approach the theoretically predicted limits we have made use of a combination of i) recent advances in semiconductor purities[3], ii) the discovery of novel donors which do not exhibit any stress dependence of their ground and excited state binding energies[3], iii) the high sensitivity of photothermal ionization spectroscopy (PTIS) and iv) magneto-optical spectroscopy using the extremely narrow lines (~10 MHz) of CO_2 laser pumped alcohol lasers.

There have been some experiments on the broadening of impurity states in Si and Ge: concentration and temperature broadening of boron lines in Si[4], the concentration dependence of the Sb lines in Ge[5], electric field-Stark broadening of impurities in Ge[6]. All these experiments were performed with electrically active impurity

concentrations larger than 10^{13} cm^{-3} and were clearly not free of stress and concentration broadenings. The best estimated value for the intrinsic FWHM of the transitions observed is ~ 50 μeV. The most recent neutron transmutation data of Jagannath et. al.[7] for P in Si appear to be substantially free of these effects, and lines were observed with FWHM ~25 μeV.

The Ge samples we have used have a donor concentration of ≤ 10^{11} cm^{-3} and have dimensions of 1x3x7 mm^3. Ion implanted ohmic contacts[3] were applied. The photothermal conductivity signal (PTCS) generated by far-infrared (FIR) laser radiation is monitored with the sample subjected to a sweeping magnetic field parallel to the [111] direction. The unpolarized incident light had E ⊥ [111]. The FIR laser radiation lines used were λ=117.227, 115.823, 108.74, 103.481, and 103.125 μm. The actual H field on the sample was measured with an uncertainty of up to 5%. It has a 2% inhomogeneity in the whole volume of the sample. The sample was mounted in a stress-free arrangement[3].

The Zeeman diagram for the D(H,O) donor is shown in Fig. 1. These data were used to identify the transitions which were tuned with the magnetic field to the laser frequencies[8]. Here the subscripts -A, - B denote the transitions corresponding to the valley parallel to the field and the other three equivalent valleys, respectively. Fig. 2 shows the results of the photothermal conductivity signal as a function of the magnetic field, for a laser wavelength of λ=115.823 μm. The figure shows the comparison between the recorded lineshapes for the $1S \rightarrow 2P_{-A}$ and $1S \rightarrow 2P_{-B}$ for three parameters. These are: a) band edge illumination; curve A, light on, B, light off. The band gap light provides free carriers which neutralize the ionized impurities

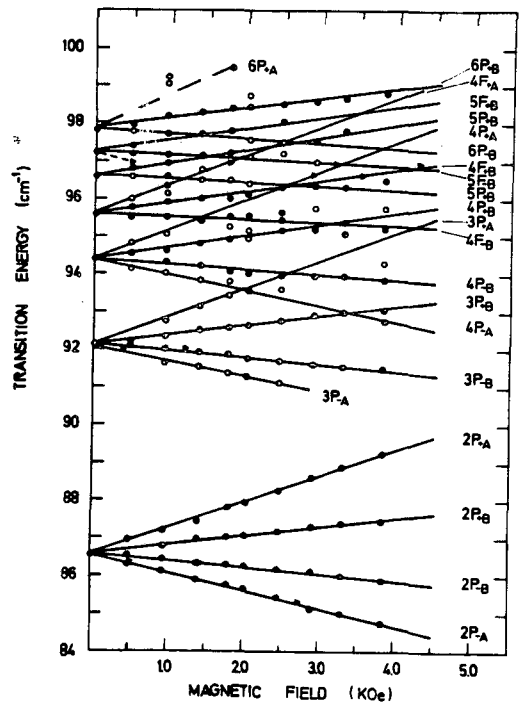

Fig. 1 The Zeeman diagram of the D(H,O) donor transition lines, measured with a Fourier Transform Interferometer.

and thus the random electric fields. Because we observe no effect on the FWHM the Stark effect must be negligible. In 2b one can see the comparison of the effect of the applied electric field E used to record the PTCS. Curve A: E=1.47 V/cm and B, E=-0.020 V/cm. Again no effect is visible in the FWHM. Fig. 2c shows that there is also no measurable effect on the FWHM for sample temperatures between 4.5 and 9 K. The average FWHM recorded for the $1S \rightarrow 2P_{-A}$ is 8.2 ±0.4 μeV. For the $1S \rightarrow 2P_{-B}$ the value is 11.4 ±0.8 μeV. The latter is broadened because of a small misalignment from the [111] direction.

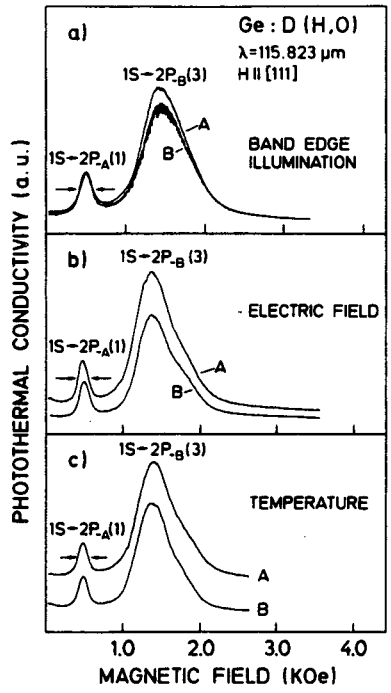

Fig. 2 Phothermal conductivity spectra of the D(H,O) donor as a function of the magnetic field. The symbols (1) and (3) denote the number of equivalent valleys with respect to H.

We will now review the contributions to the FWHM arising from various broadening mechanisms[7,9] and as estimated in ref.[10].

(I) The radiative recombination lifetime broadening[10] is expected to be ≤2.88 x 10^{-9} μeV (~.50 sec)

(II) Residual strains: The contribution due to dislocation lines (DL) for a stress sensitive center results in ~ 3 μeV for DL ~10^2 to 10^3 cm^{-2}. However the D(H,O) is one of the stress insensitive donors in Ge and no effect is expected.

(III) Electric field broadening: The combined Stark effects are estimated to give[10] a contribution ~ 2x10^{-4} μeV.

(IV) Concentration broadening. At the low concentrations of donors present no broadening is expected for the examined transitions.

(V) Bound Carrier-Phonon Interaction. The lattice vibrations broaden the electronic transitions by the finite value of the electron-phonon interaction. The $\vec{k}$ conservation restriction for emission or absorption is no longer valid due to the localized nature of the impurity state. Thus there is an intrinsic "lifetime" broadening of the transition lines due to the recombination through emission of

acoustic phonons of the carrier from the excited state. The zero temperature broadening for a 1S to $2P_0$ transition of a hydrogenic system in a semiconductor matrix is predicted[2] to be of FWHM $\Delta\omega \leq 10^{-3}R(\hbar v/a_0)$, where v is the velocity of sound and $R=\Xi^2/(\rho\hbar a_0^2 v^3)$. ρ is the density and Ξ is the deformation potential constant[2]. An estimate of this broadening results in $\Delta\omega \sim 7$ μeV for the phonon-lifetime FWHM of this transition very close to our experimentally determined value. No additional broadening is expected[10] by raising the temperature up to 25 K. Phonon lifetime broadening thus seems to be the main physical factor determining the FWHM of transitions for donor states in Ge.

Summarizing, the FWHM measured for the observed transitions of the D(H,O) donor are of the order of 8.5 μeV or smaller. The FWHM observed can be attributed to lifetime broadening produced by the permanence time ($\tau \sim (\pi\Delta\omega)^{-1} = .16$ ns) of the carrier in a bound excited state. This time is limited by recombination to the ground state through emission of acoustic phonons.

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REFERENCES

1. Nishikawa K. and Barrie R., Can. J. Phys. 41, 1135 (1963).
2. Barrie R. and Nishikawa K., Can. J. Phys. 41, 1823 (1963).
3. Haller E. E, Hansen W. L. and Goulding F. S., Advances in Phys., 30, 93 (1981).
4. White J. J., Can. J. Phys. 45, 2797 (1969).
5. Nisida Y. and Horii K., J. Phys. Soc. Japan 26, 388 (1969).
6. Ohyama T., Phys. Status Sol. (a) 98, 373 (1980).
7. Ramdas A. K. and Rodriguez S., Rep. Prog. Phys. 44, 1297 (1981). See also: Jagannath C., Grabowski Z. W., and Ramdas A. K., Phys. Rev. B, 23, 2082 (1981).
8. Navarro H., Griffin J. and Haller E. E., to be published.
9. Kogan S. M. and Lifshits T. M., Phys. Status Sol. (a) 39, 11 (1977).
10. Navarro H., Haller E. E. and Keilmann F., to be published.

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