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INCOHERENT SATURATION STUDY OF THE SELENIUM DONOR IN AlSb

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The technique of incoherent laser saturation has been used to probe the $1s(A_1) \rightarrow 1s(T_1)$ transition of the Se donor in AlSb at liquid helium temperatures. The intensity dependence of the absorptivity is described by a cascade excitation process in which at high flux two photons promote the electron into the conduction band. The $1s(T_1)$ lifetime is determined to be between 1 and 5 psec which is about three orders of magnitude smaller than found for shallow levels in three-five compounds.

ALTHOUGH HIGH POWER LASER saturation studies of shallow levels in semiconductors show that impurity lifetimes in the range 1 nsec to 1 μsec can be obtained at low temperatures [1–4], no similar measurements on deep levels have been reported. Yet incoherent saturation measurements on such transitions should be quite informative since the data would provide some evidence as to how the increase in electron–phonon coupling constant for these localized states is balanced against the highly forbidden multiphonon decay required to relax them.

In this paper the technique of incoherent saturation [5] has been used to investigate the dynamics of the $1s(A_1) \rightarrow 1s(T_1)$ transition of the Se donor in AlSb. This transition was chosen because linear spectroscopy [6] shows that it is coincident with both the 10P(18) and 10P(20) CO_2 gas laser lines. At a sample temperature of 1.7 K the saturation of the absorptivity commences at a beam intensity of a few hundred kW cm^{-2}. Although the rate of change of absorptivity with incident intensity is much faster than can be explained by a homogeneously or inhomogeneously broadened two level system, the data can be understood with a three level cascade excitation model. The measured excited state lifetime is $\sim$1–5 psec which is about 10^3 times smaller than the values observed for shallow defects [1–4].

The sample of Se doped AlSb is from the batch described in [6]. Our spectroscopically determined values of the linewidth and center frequency of the $1s(A_1) \rightarrow 1s(T_1)$ transition as a function of temperature are shown in Fig. 1. At low temperature the transition which occurs at 944.71 cm^{-1} is straddled by two CO_2 laser lines, 10P(18) at 945.98 cm^{-1} and 10P(20) at 944.19 cm^{-1}. The temperature independence of the linewidth at low temperatures can be interpreted in one of two ways; either the line is homogeneously broadened and the low temperature linewidth is a measure of the intrinsic multiphonon decay lifetime or the line is inhomogeneously broadened due to crystal strains so the low temperature line width obtained from linear spectroscopy provides no intrinsic information about the lifetime. For the former case one can readily estimate the laser intensity where saturation should commence [5] since

$$I_s = \frac{\hbar\omega}{2\sigma}(\Delta\omega), \tag{1}$$

where ω is the center frequency, $\Delta\omega$ is the linewidth (FWHM) and σ is the absorption cross section. For this transition the linewidth is 5.6 cm^{-1} and $\sigma = 2.7 \times 10^{-15}$ cm^{-2} so $I_s = 3.7$ MW cm^{-2}. Since the saturation parameter will be smaller for the case of inhomogeneous broadening [5], the I_s calculated above represents the maximum possible value for this 2 level system.

The incoherent saturation measurements have been made with the experimental setup shown in Fig. 2. A line tunable CO_2 laser Q-switched at a frequency of 170 Hz produces 200 nsec pulses with peak power of 1.5 kW. The beam is focused to a spot size of 9×10^{-4} cm^2 on the sample which is immersed in pumped He at 1.7 K in an optical access cryostat. Sample transmission as a function of incident laser intensity is obtained by moving the CaF_2 calibrated attenuators (labeled A in Fig. 2) one by one from one side of the cryostat to the other so that errors associated with nonlinear detector response are minimized. The laser output is monitored with a pyroelectric detector which is also used to normalize the transmission by means of a two channel box car integrator.

The experimental incoherent saturation results are shown in Fig. 3. The data in the top picture are for

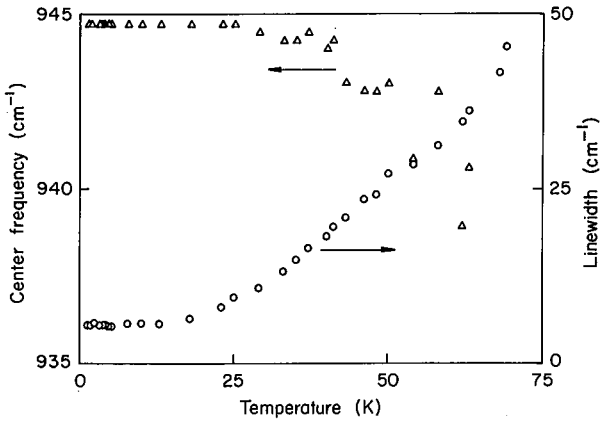

Fig. 1. Temperature dependence of the line width and center frequency of the $1s(A_1) \rightarrow 1s(T_1)$ transition in AlSb:Se. The value of the low intensity absorption coefficient, α_0, at line center is 4.2 cm^{-1}.

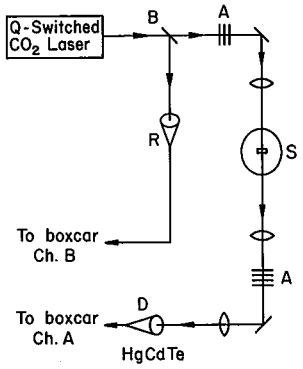

Fig. 2. Experimental arrangement for incoherent saturation measurement. Legend: B, beamsplitter; A, CaF_2 calibrated attenuators; D, HgCdTe detector; R, pyroelectric reference detector; and S, sample cryostat.

10P(18) and the bottom picture are for 10P(20). Inspection of these data show that they are qualitatively similar but quantitatively different. The saturation intensity, I_s, taken here to be equal to the incident intensity value at which the absorption coefficient is equal to one half of its low intensity value, is 120 kW cm^{-2} for 10P(18) and 400 kW cm^{-2} for 10P(20). The fact that the two curves do not look identical may be an indication of heating since with increasing temperature the absorption line center frequency in Fig. 1 moves to lower frequencies hence away from the 10P(18) line and towards the 10P(20) line. Such an effect does occur in the data shown in Fig. 3; however, the experiment described next indicates that it is electron heating and not sample heating that is important. By running the laser in a CW-chopped mode, the energy per pulse at the sample can be increased by an order of magnitude over

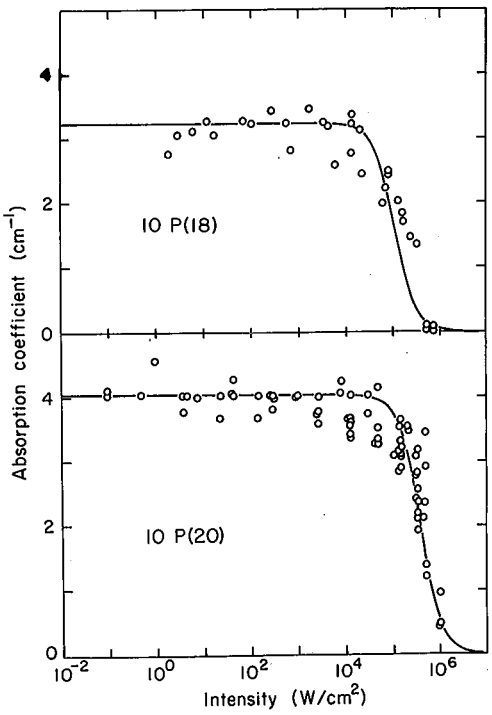

Fig. 3. Absorption coefficient vs laser intensity. Top: 10P(18) laser line on the high frequency side of the $1s(A_1) \rightarrow 1s(T_1)$ donor line. Bottom: 10P(20) on the low frequency side of the donor line. The sample temperature is 1.7 K.

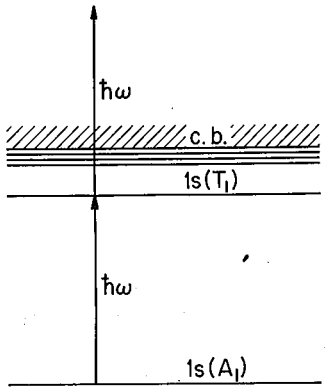

Fig. 4. Schematic diagram of the cascade excitation model.

that in the Q-switched mode. No change in sample transmission occurs in this configuration indicating that sample heating is not important. Another unusual features of the data in Fig. 3 is that the rate of decrease in absorptivity with increasing incident laser intensity is much faster than can be explained by either the homogeneous or inhomogeneous two level system [5]. Because of the electron heating contribution to the intensity dependent transmission only the 10P(20) data which is closest to line center will be analyzed below.

Both the rapid intensity dependence of the absorptivity shown in Fig. 3 and the electron heating result can be understood in terms of a three level cascade excitation model which promotes carriers into the conduction band. This idea is illustrated in Fig. 4. At high laser intensity the first $\hbar\omega$ takes the carrier from the $1s(A_1)$ to $1s(T_1)$ and the second $\hbar\omega$ takes it from $1s(T_1)$ to the conduction band. If the relaxation time from the conduction band back to the bound states is long enough then the $1s(A_1) \rightarrow 1s(T_1)$ transition will appear to saturate at high intensities but the saturation intensity will not be related to the homogeneous line width obtained from linear spectroscopy.

To estimate the functional dependence of this effect with intensity the problem is simplified to three levels. If the saturation intensity between the first two states is called I_{s_1} and the parameter for the next two states, I_{s_2}, then the steady state solution of the rate equations for the three level model gives for the absorption coefficient

$$\alpha = \alpha_0/[1 + (I/I_{s_1}) + (I/\bar{I})^2], \tag{2}$$

where

$$\bar{I} = [4I_{s_1}I_{s_2}]^{1/2}. \tag{3}$$

The effective saturation intensity in the high intensity limit is the geometric mean of the saturation intensity for each of the transitions. In this derivation it has been assumed that rapid relaxation occurs within the conduction band and also that the time for relaxation from the conduction band directly to the ground state is much larger than any of the other times in the problem. The solid curve for 10P(20) in Fig. 3 is obtained with $I_{s_1} = 3.7\,\mathrm{MW\,cm^{-2}}$ and $\bar{I} = 420\,\mathrm{kW\,cm^{-2}}$. The reasonable fit to the data demonstrates that the cascade excitation process completely masks the saturation behavior of the $1s(A_1) \rightarrow 1s(T_1)$ transition so it is not possible from this incoherent saturation measurement to determine conclusively whether or not the transition is homogeneously broadened; however, the absorption linewidth and the large intensity range for which saturation is not observed to occur provide definite bounds on the $1s(T_1)$ excited state lifetime. The value is between 1 and 5 psec.

The measured saturation intensity $\bar{I}$ together with I_{s_1} determined from the linewidth can be used in equation (3) to estimate the relaxation time for the electron to go from the conduction band to the $1s(T_1)$ excited state. The $1s(T_1)$ level ionization energy [6] of 29.4 meV ($237\,\mathrm{cm^{-1}}$) is close to the effective mass theory (EMT) ground state binding energy for AlSb of 40 meV ($323\,\mathrm{cm^{-1}}$) so the EMT value [7] of the photoionization cross section, $\sigma_2 \approx 5 \times 10^{-17}\,\mathrm{cm^{-2}}$, is used in the I_{s_2} expression, the excited state analogue of equation (1). Although a rather long relaxation time of 4 to 20 nsec is obtained, inspection of the phonon density-of-states of AlSb indicates that the phonon gap between the optic and acoustic phonon branches occurs for these transition energies [8].

In conclusion, incoherent saturation measurements on the $1s(A_1) \rightarrow 1s(T_1)$ transition show that the saturation intensity is orders of magnitude larger than previously measured for shallow states. The step function turn off of the absorptivity with increasing laser intensity is ascribed to a two step excitation process which promotes the electron into the conduction band coupled with a slow relaxation time back to the $1s(T_1)$ level. This process masks a precise determination of the $1s(T_1)$ excited state lifetime from the saturation profile; however, the value is between 1 and 5 psec, about three orders of magnitude smaller than found for shallow states in three-five compounds [4].

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