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Photoconductivity in a-SeTe

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Abstract. In-quadrature frequency-resolved photocurrent (FRPC) measurements were performed on amorphous SeTe films between 20 K and 290 K, and as a function of excitation intensity. These measurements yield lifetime distributions directly. The results show that the recombination takes place between distant pairs and that there is a continuous distribution of states within the mobility gap.

1. Introduction

Amorphous (a-) semiconductors and in particular chalcogenide glasses, such as a-Se, a-SeTe and a-As₂Se₃, are drawing a lot of attention due to their manifold applications in different fields [1, 2, 3, 4]. To optimize these materials for possible applications it is essential to understand the carrier recombination and transport mechanisms that are dominant in these materials. Modulated photocurrent (PC) measurements serve as an important tool to understand the recombination kinetics, which in turn gives information about the localized states present in a-semiconductors. However the nature of these localized states is very complicated and still open to investigation.

In this work, we measured the lifetime, temperature and excitation intensity dependence of optically modulated PC in a-SeTe.

2. Experimental details

The samples used in this work are thin films of a-SeTe prepared with thermal evaporation of 99.99992% pure Se and Te. During the thermal evaporation of the films the chamber pressure was about 10^{-7} Torr. The substrate (glass) was held at room temperature. The thicknesses of the samples were around 1 μm . Gold contacts were then evaporated on to the film in a coplanar configuration. The planar distance between the contacts was 0.4 mm. Copper wires were placed on the gold layers with silver paint.

The quadrature frequency resolved photocurrent (FRPC) response of the samples between 10 Hz and 100 kHz was measured as a function of the intensity of the excitation light and temperature. FRPC measurements were made with a lock-in amplifier (SR 530 Stanford Research System). The advantage of this frequency-locked a.c. measurement is that it rejects any stray light, dark current and any other unmodulated signal or modulated at other frequencies. Further details of the FRPC spectroscopy method are given elsewhere [5, 6, 7, 8]. The excitation source was an argon laser with a wavelength of 488 nm,

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corresponding to an energy of 2.54 eV much bigger than the optical band gap of these materials. During the measurements the sample was kept in an exchange-gas-type cryostat in which the temperature could be varied between room temperature and 20 K. The vacuum pressure of the cryostat was less than 10^{-5} Torr. The intensity of the excitation source was varied between 30 mW and $1 \mu\text{W}$ by using neutral density filters. During the high-intensity (30 mW) measurements the heating effect of the laser was controlled and it was determined that it did not have any significant heating effect on the sample.

3. Experimental results and discussion

Since the energy of the excitation light is much bigger than the optical band gap of these materials, we assume that the carriers are photoexcited between extended states and then a trap limited recombination occurs.

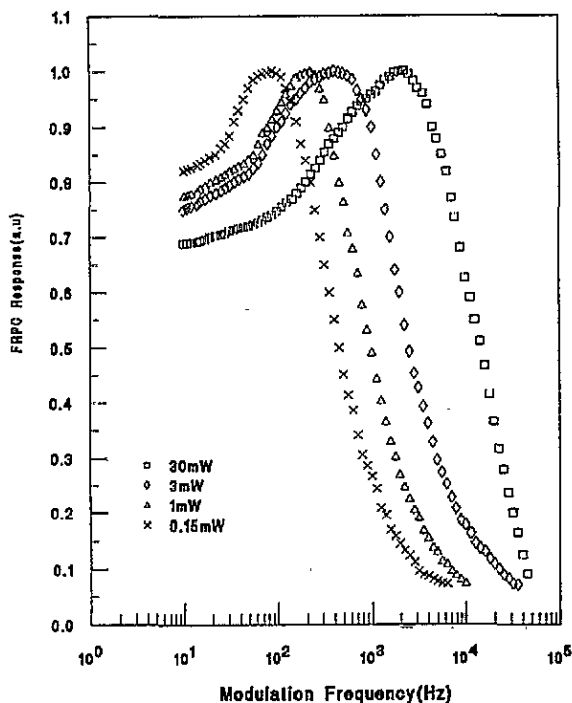


Figure 1. Frequency-resolved photocurrent response of a-SeTe at 20 K, for the indicated excitation intensities.

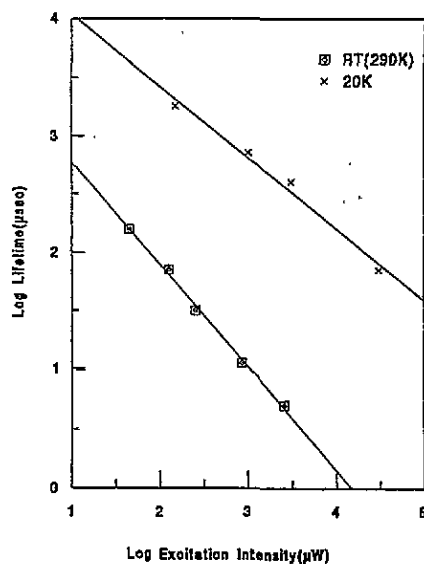


Figure 2. Photocurrent lifetime versus excitation intensity in a-SeTe at 20 K and 290 K.

Figure 1 shows the FRPC response of one of the samples for different excitation intensities at 20 K. The measurements were taken under an electric field of $1.25 \times 10^4 \text{ V cm}^{-1}$. Each curve is normalized with respect to its maximum value. As can be seen from the figure there is a single peak in each curve which shifts to higher frequencies as the intensity increases. The same measurement was repeated at room temperature (RT) and again a single peak was observed at each intensity.

The lifetime of the carriers is related to the peak frequency, f_{max} , through the relation [5]

$$\tau = \frac{1}{2\pi f_{max}} \quad (1)$$

Using these peak frequencies the lifetimes are calculated and these lifetimes are plotted against the excitation intensity in figure 2 for 20 K and room temperature. The intensity of the excitation light is proportional to the generation rate, G , and the dependence of lifetime on G can be expressed [9, 10] as

$$\tau = AG^{\nu'} \quad (2)$$

hence the slope of $\ln \tau$ against $\ln G$ will give the value for ν' . The values for 20 K and room temperature were determined to be -0.61 ± 0.04 and -0.87 ± 0.01 respectively.

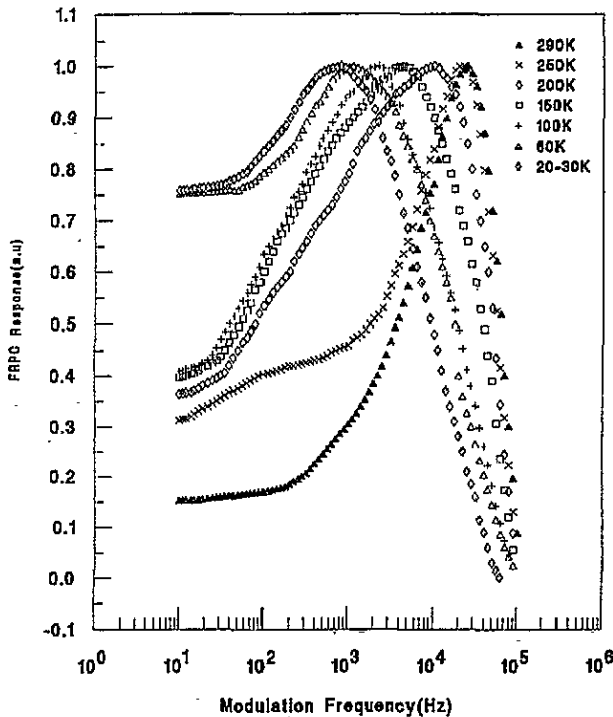


Figure 3. Frequency-resolved photocurrent response of *a*-SeTe at the indicated temperatures for an excitation intensity of 30 mW.

The distribution of lifetimes with respect to generation rate have been used to determine what kind of recombination occurs between the photoexcited carriers [11]. It has been suggested that in the geminate type of recombination the distribution of lifetimes will be insensitive to generation rates, whereas in the distant-pairs model, DP, where it is assumed that recombination takes place between nearest available neighbours non-geminately, the lifetimes should decrease with increasing generation rate. The results of figures 1 and 2 show that the lifetimes are dependent on the generation rate with a negative slope and therefore can be interpreted as supporting the DP model. Although the DP model is assumed to be valid only at low temperatures our measurements give similar results even at room temperature.

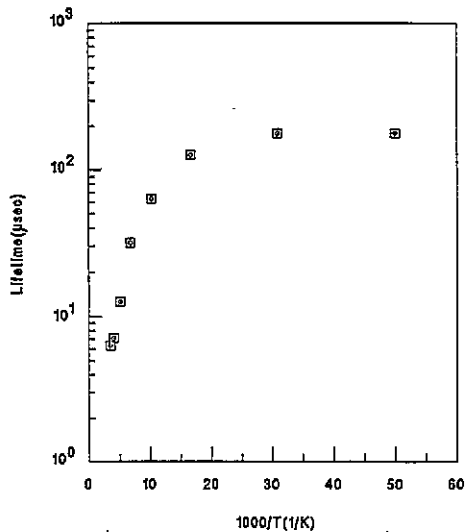


Figure 4. Photocurrent lifetime versus $1000/T$ in a-SeTe.

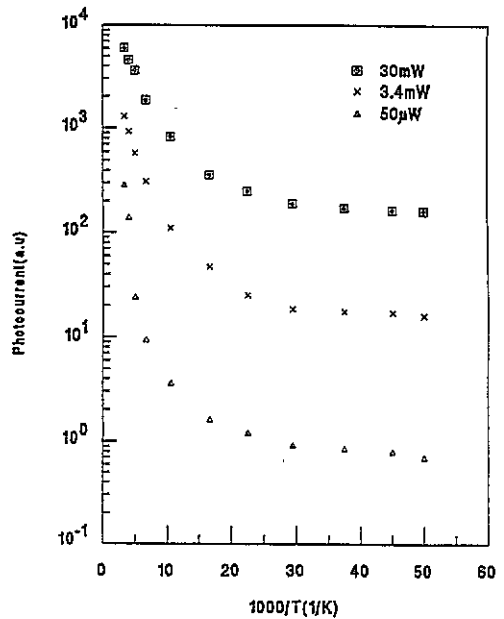


Figure 5. Temperature dependence of photocurrent in a-SeTe for three different excitation intensities measured at 10 Hz.

The frequency dependence of the FRPC response at 30 mW for different temperatures under an electric field of $2 \times 10^3 \text{ V cm}^{-1}$ is given in figure 3. Again, each curve is normalized with respect to its maximum value. This time, the peak position shifts to lower frequencies with decreasing temperatures. Using these peak frequencies the lifetimes are calculated and this time the lifetimes are plotted against $1000/T$ in figure 4. At high temperatures, between room temperature and 100 K, the behaviour of the curve is almost linear with an activation energy of 35 meV. The small value of the activation energy places the quasi-Fermi level close to the valence band, at around the lower part of an exponentially increasing tail states. The rate of increase in the lifetime decreases at lower temperatures reaching a constant value below about 60 K, indicating to the fact that at low temperatures recombination is independent of temperature.

In both the temperature- and the intensity-dependent FRPC measurements mentioned above the peak position, i.e., f_{max} , was independent of the applied electric field.

Figure 5 shows the temperature dependence of PC for three excitation intensities determined at an electric field of $1.25 \times 10^4 \text{ V cm}^{-1}$ and 10 Hz. This behaviour did not change for frequencies up to 1 kHz. In all three curves, above 200 K, the behaviour is approximately linear. For the 30 mW curve the activation energy of this linear region is about 35 meV which is in agreement with the result obtained from figure 4, confirming the position of the quasi-Fermi level. As the intensity decreases, the activation energy increases, indicating that the position of the Fermi level is moving down to deeper energy levels. At low temperatures the intensity of PC does not show an activated behaviour which is another proof that at these low temperatures the recombination processes do not depend on temperature.

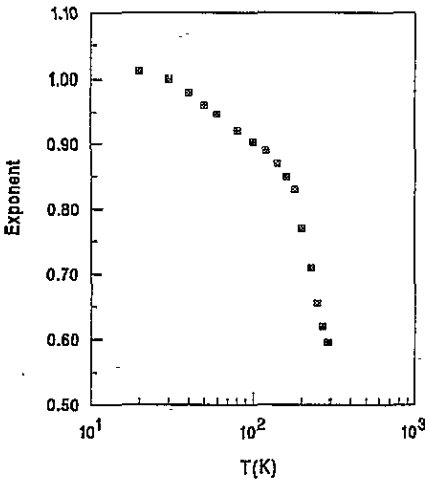


Figure 6. Temperature dependence of the exponent ν in *a*-SeTe.

The intensity of the photocurrent is proportional to the generation rate:

$$I_{PC} \propto G^{\nu} \quad (3)$$

where the exponent ν is determined to be temperature dependent. This dependence can be seen in figure 6 where the exponent calculated from $[d(\ln I_{PC})/d(\ln G)]$ is plotted against the temperature. Rose suggests that $\nu = 1$ corresponds to monomolecular recombination and $\nu = 0.5$ to bimolecular recombination [12]. However, in the case of continuous distribution of traps the value of ν may be anywhere between 0.5 and 1.0 depending upon the intensity and the temperature range. As can be seen from Figure 6 the value of the exponent lies between about 0.6 and 1.0 for the *a*-SeTe samples studied in this work, indicating the presence of a continuous distribution of localized states in the energy gap. Results for other chalcogenide samples such as *a*-Se and *a*-As₂Se₃ [8] show a constant value for ν below 100 K. This suggests that in these materials mid band gap is rather empty, i.e., there are no shallow localized states whereas they exist in *a*-SeTe samples.

4. Conclusion

The quadrature frequency resolved photocurrent response of *a*-SeTe thin-film samples between 10 Hz and 100 kHz was measured as a function of the intensity of the excitation light and temperature. The temperature range covered was 20–290 K. It was determined that the carrier lifetimes depend on the intensity of the excitation light and the temperature which may be interpreted as supportive as the DP model. The lifetimes show an activated behaviour at high temperatures, between 100 K and 290 K, with an activation energy of 35 meV for an excitation intensity of 30 mW. However, between 100 K and 20 K, the lifetimes tend to be constant. The dependence on the intensity can be expressed as $\tau \propto G^{-0.61 \pm 0.04}$ at 20 K and $\tau \propto G^{-0.87 \pm 0.01}$ at RT.

The intensity of the photocurrent at a fixed frequency is also found to be temperature dependent. The high-temperature regions of PC versus inverse temperature curves show an activated behaviour with an activation energy of 35 meV for an excitation intensity of 30 mW. For lower excitation intensities the activation energies increase due to the shift of quasi-Fermi level.

The exponent ν in the power law relationship of the intensity of the photocurrent on the generation rate varies between 1.0 and 0.6 indicating a continuous distribution of localized states within the mobility gap.

Acknowledgments

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