

Electrical properties of CdTe:Cl

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Received 11 January 1980

Abstract. The electrical properties of a series of CdTe:Cl samples cut from the same crystal boule are reported. Measurement of the thermal activation energies for both ohmic and space-charge-limited conduction reveals that the conductivity of the samples is controlled by two localised impurity levels within the band gap. A deep level at approximately 0.98 eV above the valence band edge is found in all the specimens but the energy depth of the second level is a function of the position of the sample in the crystal boule and the temperature of any annealing treatment. The conductivity of the samples also obeys the Meyer–Neldel rule or compensation law.

1. Introduction

The combination of large band gap, high average atomic number and reasonable mobility–lifetime product for both electrons and holes makes CdTe an attractive material for use in gamma radiation detectors (Miller 1972). Semi-insulating CdTe offers the additional advantage that such devices can be operated at room temperature. This material may also be useful as a substrate on which to grow epilayers of CdTe and $\text{Cd}_x\text{Hg}_{1-x}\text{Te}$.

One method of producing high resistivity CdTe is by the addition of chlorine in order to compensate for native acceptor defects. The exact compensation mechanism has not been established and to date a number of different schemes have been suggested. Chlorine was originally thought to act as a simple donor by substituting for tellurium in the lattice (De Nobel 1959). However, a number of models which consider the possibility of chlorine atoms forming complex defects have recently been put forward. A review of these ideas has been presented by Marfaing (1977).

A variety of experimental techniques has been used to determine the positions of deep traps in CdTe:Cl. Unfortunately there is still little agreement about the origin of these reported levels. Zoul and Klier (1977) have studied the temperature dependence of space-charge-limited current and interpret their results in terms of hole traps 0.40 eV and 0.65 eV above the valence band edge. These authors suggest that both the levels might be associated with a complex chlorine defect. Using time-of-flight techniques Ottaviani (1977) and Bell *et al* (1974) have identified a hole trap at $E_v + 0.35$ eV. The latter group calculate the density of this trap to be $6 \times 10^{15} \text{ cm}^{-3}$ and suggest that copper impurities might be responsible. Höschl *et al* (1975) have measured the conductivity of crystals grown under slightly different conditions and observed three groups of samples characterised by thermal activation energies of 0.15, 0.5 and 0.9 eV. The 0.15 and 0.9 eV energies are interpreted as levels above the valence band edge representing the singly and

doubly ionised cadmium vacancy respectively. The 0.5 eV energy is assigned to a cadmium vacancy–chlorine complex. More recently, from photo-Hall measurements, Höschl *et al* (1978) have reported levels at 0.55 eV and 0.90 eV above the valence band edge. Arkadyeva and Matveev (1977) have studied the Hall effect and conductivity in a number of crystals and report the most commonly occurring deep levels to be $E_v + 0.61$, $E_v + 0.73$ and $E_v + 0.83$ eV. Other experimental investigations of impurity levels in CdTe:Cl have involved thermally stimulated currents (Mancini *et al* 1977, Barnes and Zanio 1976), thermally stimulated capacitance and photocapacitance (Legros *et al* 1977), photoluminescence (Agrinskaya and Matveev 1976) and electroabsorption (Neu *et al* 1977).

In this work a systematic investigation of the conductivity of a series of CdTe:Cl samples taken from one crystal boule is reported. The results are interpreted using theorems proposed by Roberts and Schmidlin (1969) and Schmidlin and Roberts (1974).

2. Experimental

The measurements reported in this paper were made on a series of CdTe:Cl specimens sliced from a single boule. These crystals were grown at RSRE, Malvern, using the solvent evaporation technique (Lunn and Bettridge 1977). The slices of crystal ($\langle 111 \rangle$ orientation and typically 1000 μm thick) were mechanically polished to a finish of 1 μm using diamond paste and were then chemically polished in a solution of 1% bromine in methanol. Before evaporation of contacts the samples were etched in 40% hydrofluoric acid for 1 min. This was necessary to give reproducible injecting contacts. A similar treatment has been found to leave a Te-rich surface on p-type CdTe (Montgomery 1964). Contacts of bismuth or gold were evaporated on to opposite faces of the samples at a pressure of 10^{-6} Torr. Conductivity measurements over the temperature range 200–370 K were made in a helium exchange gas cryostat using a Keithley 241 regulated power supply and 410 A picoammeter.

3. Results and discussion

3.1 Nonextrinsic conduction

All the samples investigated exhibited ohmic conductivity at low applied fields. At electric fields greater than about 10^5 V m^{-1} space-charge-limited conduction was observed, being characterised by a square law dependence of current upon applied field. Similar results have been reported previously for undoped CdTe (Pande and Roberts 1977). Figure 1 shows the temperature dependence of the ohmic and space-charge-limited current for one of the samples. In the temperature range investigated there is one activation energy (0.62 eV) for the space-charge-limited current, whereas the ohmic current shows two distinct energies (0.62 eV, 0.80 eV) with a transition temperature of 256 K. These different activation energies were interpreted using a theorem proposed by Roberts and Schmidlin (1969). This states:

‘The existence of different (or identical) activation energies for Ohmic and SCL conduction is both a necessary and a sufficient condition for Ohmic conduction to be non-extrinsic (or extrinsic).’

The conductivity of a material is referred to as nonextrinsic (or pseudointrinsic) when it is dominated by two localised levels within the band gap. From figure 1, the ohmic

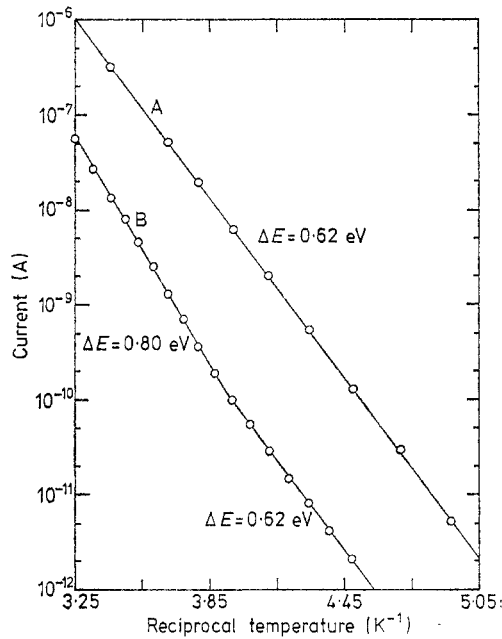


Figure 1. Current versus reciprocal temperature in the ohmic and space-charge-limited regions for a CdTe:Cl sample. Thickness=1.5 mm. Electrode area=7 mm². A, space-charge-limited current; B, ohmic current.

conduction in our CdTe:Cl sample is extrinsic at temperatures up to 256 K and non-extrinsic above this.

In order to interpret in detail the electrical conductivity data for the CdTe:Cl samples, the conductivity type must be known. Unfortunately, many of our samples were too resistive for simple Hall-effect experiments. However, because ohmic contacts could be made to the specimens using high work-function metals (e.g. Au) and Schottky barriers were produced using low work-function metal electrodes (e.g. In), it was concluded that the material was probably p-type. This agrees with other authors (e.g. Strauss 1977). The activation energy for extrinsic conduction is therefore contained in the following equation for p , the concentration of holes in the valence band:

$$p = [(N_a - N_d)/N_m] N_v \exp [(E_m - E_v)/kT]. \quad (1)$$

N_a , N_d are the concentrations of shallow (ionised) acceptors and donors respectively, N_v is the density of states in the valence band, $(E_m - E_v)$ is the energy separation between the dominant hole level and the valence band (a negative quantity for the sign convention adopted throughout this paper), and N_m is the concentration of this level. For extrinsic conduction the Fermi level must be positioned so that the total number of holes, p_m , in the level E_m is equal to $(N_a - N_d)$.

In the nonextrinsic region the ohmic conductivity is determined by an additional level E_q , of concentration N_q . E_q is the dominant electron level and may be considered as a deep acceptor. For nonextrinsic conduction the Fermi level will be positioned between levels E_m and E_q so that the total number of holes, p_m , in level E_m is equal to the total number of electrons, n_q , in level E_q . The activation energy for nonextrinsic conduction is now contained in the expression (Roberts and Schmidlin 1969):

$$p = N_v (N_q/N_m)^{1/2} \exp \{[(E_m - E_v) - 0.5 (E_m - E_q)]/kT\}. \quad (2)$$

Note that it is possible for the dominant hole and electron levels to be the valence and conduction bands respectively, in which case $E_m = E_v$, $E_q = E_c$ and equation (2) reduces to the familiar form for intrinsic conduction.

The activation energy for space-charge-limited conduction in both the extrinsic and nonextrinsic cases is given by the following equation for θ_p , the fraction of total carriers (all holes below the Fermi level) which are free:

$$\theta_p = (N_v/N_m) \exp [(E_m - E_v)/kT]. \quad (3)$$

θ_p is related to the space-charge-limited current density, J_{sc} , by the equation:

$$J_{sc} = \frac{2}{3} \theta_p \epsilon \mu_p (V^2/L^3) \quad (4)$$

where V is the voltage across a sample of thickness L , μ_p is the hole drift mobility and ϵ is the permittivity.

Using the above equations and the data from figure 1 it can be seen that the ohmic conductivity in the sample is governed by two levels, $E_m - E_v = 0.62$ eV and $E_q - E_v = 0.98$ eV. At temperatures below 256 K the conduction is extrinsic and the activation energy is simply 0.62 eV. Above 256 K the conduction becomes nonextrinsic with an activation energy of $0.98 - 0.5$ ($0.98 - 0.62$) = 0.80 eV.

In order to obtain information about the densities of the levels E_m and E_q and to explain the transition from extrinsic to nonextrinsic conduction, the following procedure was adopted:

- (i) Using the values of the space-charge-limited and ohmic currents for extrinsic conductivity, the value of the excess acceptor concentration ($N_a - N_d$) was calculated via equations (1), (3) and (4).
- (ii) A value for μ_p , the hole mobility, was assumed and values for N_m and N_q were derived using the graphical approach suggested by Schmidlin and Roberts (1974).
- (iii) Finally an iterative routine on a computer was used to 'fine-tune' the above values so as to obtain an exact fit with the experimental data. The computer programme was a general one using Fermi-Dirac statistics and simply evaluated the position of the Fermi level from the charge neutrality equation for a selected impurity level distribution. The following parameters were used: $E_v - E_c = 1.61 - (3 \times 10^{-4})T$ eV, where T is the temperature in K (Strauss 1977, Camassel *et al* 1973).

$$\left. \begin{aligned} E_v - E_m &= 0.62 - (0.62/1.61) \times 3 \times 10^{-4} T \text{ eV} \\ E_v - E_q &= 0.98 - (0.98/1.61) \times 3 \times 10^{-4} T \text{ eV} \end{aligned} \right\} \begin{aligned} m_e^* &= 0.14 m_0 \\ m_p^* &= 0.35 m_0 \\ \mu_e &= 1500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \\ \epsilon &= 11 \epsilon_0. \end{aligned} \quad (\text{De Nobel } 1959)$$

The assumption that the energy separations of the deep levels from the band edges remain proportional to the energy gap as it changes with temperature is probably an oversimplification. However, lacking detailed information about deep levels in CdTe, this is the best approximation we can make. Other workers have used a similar approach (e.g. Zucca 1977). The hole mobility, μ_p , was assumed to have a temperature dependence of the form $\mu_p(T) = \mu_p(0)T^{-1.5}$. No allowance was made for the spin degeneracy of levels E_m and E_q in the iteration. The best computer fit was obtained using the values:

$$\begin{aligned} N_m &= 1.00 \times 10^{12} \text{ cm}^{-3} \\ N_q &= 2.00 \times 10^{15} \text{ cm}^{-3} \\ N_a - N_d &= 2.40 \times 10^{10} \text{ cm}^{-3} \end{aligned} \quad \begin{aligned} \mu_p(0) &= 85\,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \\ (\text{i.e. } \mu_p(293) &= 169 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}). \end{aligned}$$

The results are shown in figure 2 in a graphical form in which log of state density is plotted against position in the band gap. The apex of the 'V', whose sides have slope $\pm(\lg e)/kT$, lies on the abscissa at the Fermi energy. The vertical extension of any state density, N_i , above the 'V' gives the population of that state. Thus the vertical extensions of state densities above the left side of the 'V' represent the population of those states with holes while the vertical extensions of the state densities above the right side of the 'V' represent the population of those states with electrons.

Figure 2(a) shows the situation at 220 K (extrinsic region). The dominant hole level is E_m and the Fermi level is positioned just above this level in the band gap so that $(N_a - N_d) = p_m$. As the temperature increases the Fermi level must move to maintain this

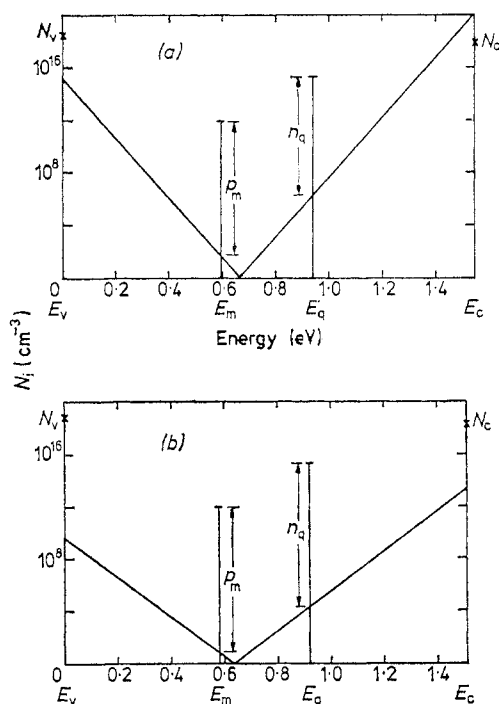


Figure 2. Graphical representation of the state density as a function of energy for CdTe:Cl. The values of position and density of the levels are 'best fit' computed values for the sample whose conductivity characteristics are shown in figure 1. (a) 220 K, extrinsic. $N_a - N_d = p_m$. (b) 330 K, nonextrinsic. $p_m = n_q$.

condition of charge neutrality. However, the population of electrons in E_q increases with temperature and the carrier statistics will begin to become dominated by this level. At a certain temperature the concentration of electrons, n_q , in E_q will become equal to the concentration of holes, p_m , in E_m . The conductivity then becomes nonextrinsic with a resultant increase in the ohmic activation energy of $0.5 (E_m - E_q)$ (Schmidlin and Roberts 1974). The situation at 330 K, well above this transition temperature, is shown in figure 2(b).

Figure 3 (full curve) shows the computed Fermi level movement over the temperature range of the measurements. In the extrinsic conduction region the Fermi level moves away from the valence band as the temperature is increased to maintain the charge

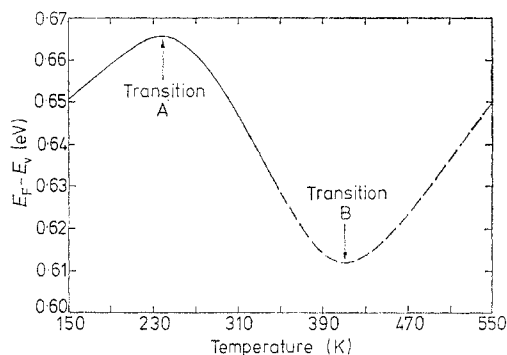


Figure 3. Computed Fermi level position (measured from the valence band edge) with temperature for the CdTe:Cl sample whose conductivity characteristics are shown in figure 1.

neutrality condition. At about 240 K (transition A) the direction of Fermi level shift with temperature changes as the conduction becomes nonextrinsic. At higher temperatures (broken curve) it is expected that the Fermi level would change direction again (transition B) as dominance is shifted from E_m to E_v and from E_q to E_c . The conductivity would then be intrinsic. For reasons given in §3.3 we could not heat our samples to the required temperature to observe this second transition.

3.2. Meyer-Neldel rule

All of the CdTe:Cl samples investigated showed activation energies for ohmic and space-charge-limited currents qualitatively similar to those in figure 1, i.e. in the temperature range 200–370 K, two activation energies were observed for ohmic conduction and one such energy was observed for space-charge-limited conduction. However, the exact values of these activation energies and the transition temperature from extrinsic to non-extrinsic conduction varied from one sample to another. Following the interpretation discussed in section 3.1 for each set of results, we discovered that the deep E_q level remained in approximately the same position in the energy gap (0.98 eV above the valence band edge) for different samples whereas the position of the E_m level varied considerably. An estimate of state densities, using the method outlined in the previous section, also indicated that the concentration of the E_m level remained constant in different samples, but that the concentrations of both the E_q level and the excess acceptors were changed.

Figure 4 shows the ohmic activation energies in the extrinsic conduction region (i.e. simple ionisation energies) for three samples sliced from different positions in the CdTe:Cl crystal boule. Thus the depth of the level E_m from the valence band edge varies from 0.32 eV to 0.83 eV for different samples (in general the more resistive samples were cut near the seed end of the ingot). From our experiments it was difficult to determine whether there existed a continuous series of activation energies in different crystals or whether there was a finite number of discrete levels. As can be seen the conductivity versus reciprocal temperature curves meet at a focal point ($T=478$ K, $\sigma=5 \times 10^{-4} \Omega^{-1} \text{ m}^{-1}$) when extrapolated to higher temperatures. A similar set of curves was obtained when the ohmic conductivity in the nonextrinsic region was plotted as a function of reciprocal temperature. In this case the curves, when extended, passed through a focal point at $T=806$ K, $\sigma=16 \Omega^{-1} \text{ m}^{-1}$.

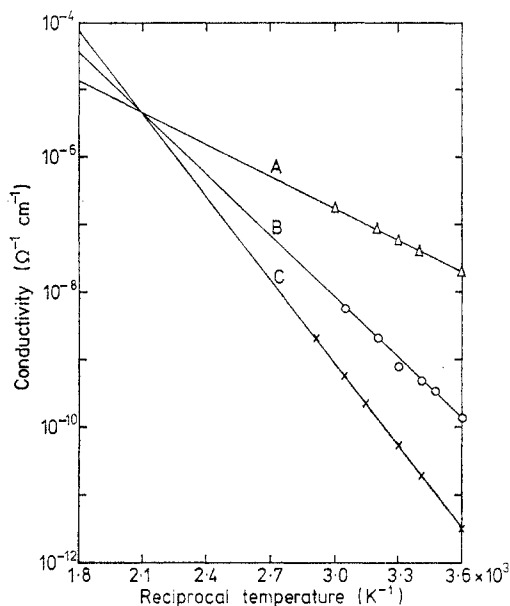


Figure 4. Ohmic conductivity in the extrinsic region versus reciprocal temperature for three CdTe:Cl samples cut from the same crystal boule. Characteristic temperature 478 K. A, $\Delta E=0.32$ eV; B, $\Delta E=0.58$ eV; C, $\Delta E=0.83$ eV.

Graphs similar to those in figure 4 are often observed for semi-insulating materials, particularly organic semiconductors. This behaviour was first noted by Meyer (1933); subsequently Meyer and Neldel (1937) showed that there was a linear relationship between the slopes and intercepts of the conductivity Arrhenius plots for various metallic oxide samples. This may be written

$$\ln \sigma_0 = \alpha E + \beta \quad (5)$$

where α, β are constants and σ_0, E are parameters in the Arrhenius relationship for conductivity

$$\sigma(T) = \sigma_0 \exp(-E/kT). \quad (6)$$

Equation (5) is often referred to as the Meyer-Neldel or compensation law.

Rosenberg *et al* (1968) showed that equation (6) was incomplete in the case of activation energy curves intersecting at a focal point $T=T_c$ and not infinity. To explain their results these authors suggested a three-constant equation of the form

$$\sigma(T) = \sigma_0' \exp(E/kT_c) \exp(-E/kT). \quad (7)$$

The additional constant, T_c , was referred to as the characteristic temperature. Comparing equations (5), (6) and (7) shows that $\alpha = (kT_c)^{-1}$ and $\beta = \ln \sigma_0'$. Rosenberg *et al* (1968) have tabulated results for a number of organic substances and show T_c to vary from 270 K to infinity. They suggest that this parameter is a molecular property of the material and is independent of the crystal structure and the nature of the electrodes.

Although there have been several attempts to explain the Meyer-Neldel rule for specific experimental data (Nespurek *et al* 1976, Dewsberry 1976), to date there is no general theory for this empirical relationship. Roberts (1971) has discussed the Meyer-

Neldel rule in terms of nonextrinsic conduction. This author considered a solid which is subjected to a disturbance so that the statistical distribution of carriers in the energy gap is changed, and derived expressions relating the characteristic temperature to the state densities and energy levels before and after the disturbance. The analysis assumes that the carrier mobilities are unaffected by this disturbance. As an example, suppose a level E_{m1} of density N_{m1} (in a p-type solid) is replaced as the dominant hole level by a state with density N_{m2} and energy E_{m2} . At the same time, although the dominant electron level remains at the same energy, its density is altered to N_{q2} . The conductivity versus reciprocal temperature curves before and after the disturbance will then intersect at $T = \infty$ provided

$$N_{m2}/N_{m1} = N_{q2}/N_{q1}. \quad (8)$$

Part of this model seems acceptable for our CdTe:Cl samples since we observe that only the E_m level changes from sample to sample while the E_q level remains in approximately the same position in the energy gap. However, because the extrinsic and non-extrinsic conductivity versus reciprocal temperature curves have focal points at finite temperatures, equation (8) cannot provide a complete explanation for the occurrence of the Meyer-Neldel rule. For the curves to intersect at a finite temperature imposes a considerable constraint on the relative values of the sample parameters. For example, following the approach of Roberts' (1971) paper, the following condition is obtained for a focal point to be observed in the case of non-extrinsic conduction:

$$N_{m2}N_{q1}/N_{m1}N_{q2} = \exp [(E_{m2} - E_{m1})/kT_{cn}]. \quad (9)$$

T_{cn} is the characteristic temperature for nonextrinsic conduction (806 K for our material). Note that the sign convention for the energy levels in equation (9) is the same as that adopted in equations (1), (2) and (3), i.e. if E_{m2} is closer to the valence band than E_{m1} , then $E_{m2} - E_{m1}$ is a positive quantity. Using equation (1) a similar expression can be obtained for the case of extrinsic conduction. The restrictions imposed by such equations on the densities and energies of the impurity levels in the CdTe would seem to preclude the possibility that these parameters are completely independent quantities. Hence it is likely that a relationship exists between these quantities; this is further discussed in §3.4.

3.3. Annealing effects

If the CdTe:Cl crystal slices were heated to temperatures greater than 120 °C, irreversible changes in the electrical properties were observed. In particular, the conductivity curves displayed smaller activation energies as the annealing temperature was increased. If the activation energy data for one sample before and after various annealing treatments were plotted on the same graph a 'fan' of curves, similar to that in figure 4, was obtained for both extrinsic and nonextrinsic conduction. The Meyer-Neldel rule would, therefore, seem to apply to one CdTe:Cl sample under different annealing conditions as well as to different samples from the same crystal boule. Figure 5 shows the effect of annealing on the extrinsic activation energy for one sample. In this graph an alternative method of demonstrating the Meyer-Neldel relationship is used from which the validity of equation (5) is clearly shown. The characteristic temperature, calculated from the slope of the graph, is 481 K, which compares favourably with the value of 478 K obtained for different un-annealed samples. A similar exercise for the nonextrinsic activation energies gave a

characteristic temperature of 806 K (exactly the same value obtained for different samples).

In order to check that we were not observing an annealing effect of the electrical contacts the following experiment was performed on a number of samples; after heating the sample at a temperature of about 300 °C the thermal activation energies for ohmic conduction were obtained. The contacts were then etched away and new electrodes deposited on to the crystal slice. Finally, the ohmic activation energies were remeasured. This experiment always produced identical results with the 'old' and the 'new' contacts inferring that the annealing treatment had affected the bulk of the CdTe:Cl crystal rather than the electrodes or the surface.

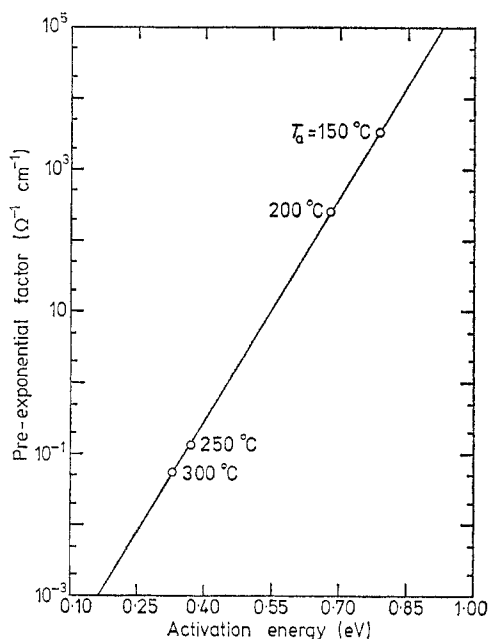


Figure 5. Pre-exponential factor versus ohmic activation energy for extrinsic conductivity of one CdTe:Cl sample after annealing in vacuum for 20 min at different temperatures (T_a). Characteristic temperature 481 K.

Interpretation of the activation energy curves after annealing using the theories of Roberts and Schmidlin revealed, once again, that the energy position, E_m , of the dominant hole level was changing under different annealing conditions while the dominant electron level at E_q remained in the same position in the energy gap. In fact for annealing temperatures below 300 °C the E_m level varied from about 0.8 eV to 0.3 eV above the valence band edge. This was approximately the same variation observed in different (unannealed) samples (see figure 4). If the samples were annealed at temperatures greater than 300 °C a constant extrinsic activation energy of 0.30 eV was obtained. If the annealing process was continued for about 3 h this activation energy was reduced to 0.15 eV. The conductivity data in these cases did not lie on the straight line shown in figure 5 but the pre-exponential factor continued to decrease with increasing annealing temperature.

3.4. Origin of deep levels

From the experimental data presented in this paper it is difficult to accurately assign the energy levels observed to specific defects in the CdTe. However, some tentative conclusions may be drawn. We note that the E_q (deep acceptor) level is observed in approximately the same position in the band gap (0.98 eV above the valence band edge at 0 K) in all our samples. One possible interpretation would be to assign this level to a doubly ionised cadmium vacancy, which is the most frequently reported deep acceptor-type defect in CdTe:Cl. The position of this defect is given by many authors to be in the range 0.65–0.9 eV above the valence band edge (Strauss 1977, Marfaing 1977, Kroger 1977, Höschl *et al* 1977). The 0.15 eV activation energy, observed after annealing the crystals above 300 °C, has also been reported by the same authors but there is no agreement on its origin.

The explanation of the variable E_m (deep donor) level is more difficult. It would seem likely that this is associated with the chlorine because we have not observed such a variable level in undoped CdTe crystals grown under similar conditions (Pande and Roberts 1977). Current models for compensation in CdTe:Cl favour the presence of a complex defect in which chlorine is involved (Marfaing 1977, Legros *et al* 1978). The energy level of such complexes will, of course, depend on the degree of association of the constituent defects. Höschl *et al* (1977) have shown that a cadmium vacancy–chlorine complex can give rise to a series of discrete energy levels within the band gap of CdTe. Therefore such a defect might form the basis for an explanation of the Meyer–Neldel rule behaviour discussed in §§3.2 and 3.3. An alternative explanation might be that the presence of chlorine in CdTe gives rise to a multiplicity of levels associated with electronic states of different valency. Clearly the level which would have most influence over the position of the Fermi level will be a function of r , the ratio of the total number of electrons associated with the defect states to the number of defects. A possible explanation for the different activation energy curves observed in our CdTe:Cl samples would then be that r is a function of position in the crystal boule and of subsequent heat treatment. If either of the above suggestions is correct, this would imply that the level E_m , observed in our material, does not take a full range of values, but changes in discrete steps.

4. Summary

The results of an activation energy analysis for both ohmic and space-charge-limited current in CdTe:Cl crystals show that the conductivity is controlled by two levels in the band gap. A deep level, 0.98 eV above the valence band edge, is found in all of our samples and may possibly be attributed to the doubly ionised cadmium vacancy reported by other workers. The depth of the second level controlling the conductivity of our samples is found to depend on where the specimen is cut from the crystal boule and the temperature of any annealing treatment. We suspect that this energy level is associated with the chlorine impurity, although other investigations are necessary to confirm this. The conductivity of our samples is found to obey the Meyer–Neldel rule. This would seem to imply that there is a fundamental relationship between the concentrations and energy depths of the dominant electron and hole levels in the material.

Work is in progress to incorporate the simple multivalent picture envisaged in §3.4 into the Roberts–Schmidlin dominant level approach to activation energy analysis. We find that it is possible to calculate the Fermi level position for the simple multivalent

pair of levels without resort to the grand partition function but unfortunately the complete problem still cannot be resolved without a computer. We are currently developing the theory in order to establish whether it can provide a general account for the Meyer–Neldel rule and more specifically, whether it can explain our results for CdTe:Cl.

Acknowledgments

The authors wish to thank RSRE, Malvern and JB Mullin in particular for supplying the CdTe crystals and for useful discussions.

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