

Electrical properties of liquid Cd-Te alloys

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The electrical resistivity and the thermopower of liquid $\text{Cd}_x\text{-Te}_{1-x}$ have been investigated experimentally as a function of temperature over the whole composition range. Due to the high melting point of $\text{Cd}_{0.5}\text{Te}_{0.5}$ (1098 °C) and the low boiling points of cadmium (765 °C) and tellurium (990 °C), the measurements were difficult. Nevertheless, with a different experimental setup we were able to measure the resistivity and the thermopower from the liquidus up to 1200 °C. The resistivity shows a maximum at $x=0.54$ reaching a value of 13 000 $\mu\Omega\text{ cm}$ at 1100 °C. A marked feature of liquid $\text{Cd}_x\text{-Te}_{1-x}$ is that the thermopower varies rapidly from a positive value in the tellurium rich region to a negative value in the cadmium rich one around the equiatomic composition. The electronic properties, near this composition, were analyzed by using equations derived from the Kubo-Greenwood formula.

I. INTRODUCTION

In recent years, several measurements have been reported¹⁻³ concerning the electrical and other properties of liquid tellurium-based alloys because of their interesting behavior such as compound formation at a particular composition and the “*p-n* transition” from a semiconducting state to a metallic one with increasing temperature. But properties of II-VI compounds, in the liquid state, have not received much attention, due to the experimental difficulties coming from their high vapor pressures and high melting temperatures.

Cadmium tellurides are known as important materials for the fabrication of gamma-ray detectors. This is due to their large atomic number and their high band gap at room temperature. The nature of bonding in these compounds has been the subject of many speculations. Recently, Prigent *et al.*⁴ and Gaspard *et al.*⁵ carried out neutron-diffraction measurements on liquid $\text{Cd}_x\text{-Te}_{1-x}$ and showed that liquid $\text{Cd}_{0.5}\text{Te}_{0.5}$ keeps its fourfold coordination, which corresponds to an extension of the semiconducting state across the melting point. This is in contradiction with the more close-packed structures of liquid III-V alloys. Coordination numbers in these systems change from 4 in the crystalline phase (semiconducting) to ~ 6 in the liquid phase⁶ (metallic). It is interesting to study in details, as a function of composition, the liquid Cd-Te system which exhibits a transition between a liquid metal (cadmium or tellurium) and a liquid semiconductor ($\text{Cd}_{0.5}\text{Te}_{0.5}$).

In this paper, we present the electrical resistivity (ρ) and thermopower (S) of liquid $\text{Cd}_x\text{-Te}_{1-x}$ over the entire composition range (x). We analyze the electrical conductivity ($\sigma = 1/\rho$) and S near $x=0.5$, assuming an energy dependent conductivity⁷ $\sigma(E)$. The three regimes of σ (metallic, semi-metallic and nonmetallic) often referred to in the literature (Cutler⁸) represent a useful framework for our discussion.

II. EXPERIMENTAL PROCEDURE

For the discussion of electronic transport properties of liquid $\text{Cd}_x\text{-Te}_{1-x}$ it is necessary to know whether electrons or holes predominate. To obtain this information, mostly measurements of S are used, because its sign is closely related to the change of the dominating kind of carriers. The T

dependence of S together with that of ρ gives further information about the transport mechanism.

The resistivity as well as S have been measured using a homemade apparatus that permits to measure both at the same time. This method has been completely described by Vinckel *et al.*^{9,10} It combines a measurement of ρ by the four-point technique, and of S by employing a small ΔT method that suppresses the errors due to small variations of the thermoelectric properties of the reference wires. The liquid alloy is contained in a silica cell and the measurement are made using two tungsten and two tungsten-rhenium (W-26% Re) electrodes. Previous calibration of the tungsten reference electrodes as well as of the tungsten-rhenium counter-electrodes has been made with calibrated “platinum 67” wire, using the results of Roberts *et al.*¹¹⁻¹³ The tungsten calibration has also been compared with the values of Roberts *et al.*,¹⁵ and we obtained a maximum difference of 0.2 $\mu\text{V/K}$ at 1200 °C.

The geometrical constant of the cell, that correlates the resistance with ρ , has been carefully calibrated with triple distilled mercury at room temperature. We do not correct ρ from the thermal expansion of the silica because this effect (0.02%) is much smaller than the total experimental uncertainty (0.1%). The potential drop across the cell capillary was measured twice by reversing the current to eliminate small thermoelectric voltages.

Cadmium (Cd) and tellurium (Te) were supplied by Johnson Matthey Co. and were 99.999% pure. Measurements were carried out in an argon gas atmosphere. Because of the high vapor pressure and low boiling points of both elements, it is necessary to apply pressure to avoid the formation of bubbles inside the liquid. Special care has been taken for the Cd-rich compositions. The 9 bars pressure that we applied allowed us to measure liquid $\text{Cd}_x\text{-Te}_{1-x}$ up to 1200 °C.

III. EXPERIMENTAL RESULTS

A. Resistivity

Measurements were made on 21 different compositions. Figure 1 shows ρ and Fig. 2 shows S both as a function of T .

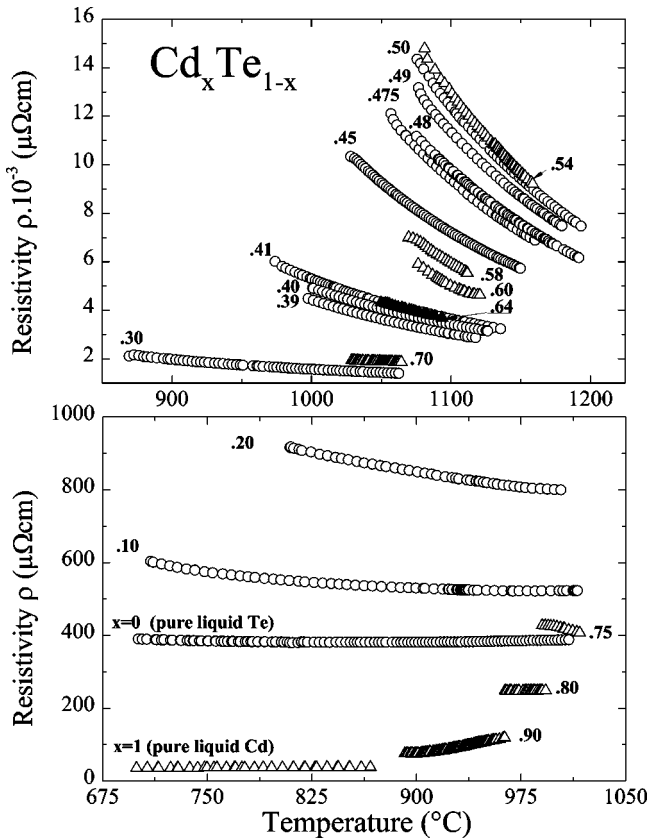


FIG. 1. Electrical resistivity as a function of temperature for liquid $\text{Cd}_x\text{Te}_{1-x}$.

For the Te-rich side ($0 \leq x \leq 0.2$) liquid $\text{Cd}_x\text{-Te}_{1-x}$ has a relatively low resistivity and a negative temperature coefficient (NTC). This negative coefficient becomes smaller at higher T . For pure Te, the temperature coefficient becomes even positive above 850°C .

For alloys with $0.30 \leq x \leq 0.54$, ρ increases with x towards the equiatomic stoichiometry and decreases with T . It is very sensitive to x and T . One finds a pronounced maximum at $x = 0.54$ with $\rho \approx 13\,000 \mu\Omega \text{ cm}$ at 1100°C . It was assumed that a conductivity gap^{14,15} $\Delta E_G = E_C - E_V$ remains in the liquid state (E_C and E_V are the conductivity edges of conduction and valence bands, respectively). Near the maximum of ρ , we observe the largest NTC. To our knowledge, there exist only resistivity measurements near the equiatomic composition (Glazov *et al.*,¹⁶ 1969). Although they claimed to have measured the equiatomic alloy, they reported a melting point of about 1050°C . Massalski *et al.*¹⁷ indicates a value of 1098°C . Due to the low melting point observed ($\sim 1050^\circ\text{C}$), it could be that Glazov did not reach the equiatomic composition. His resistivity measurement corresponds almost to our experimental data for $x = 0.48$.

For $0.58 \leq x \leq 0.75$, ρ now decreases with x and T . Prigent *et al.*⁴ showed that this composition range exhibits an intermediate coordination number and a structure different from that of a liquid metal. For liquid $\text{Cd}_{0.8}\text{Te}_{0.2}$ ($x = 0.8$), ρ is almost temperature independent and reaches a value of about $280 \mu\Omega \text{ cm}$ at the melting point which corresponds roughly to the limit between metallic and semimetallic liquid alloy.⁷ Liquid $\text{Cd}_{0.9}\text{Te}_{0.1}$ has a positive temperature coefficient of

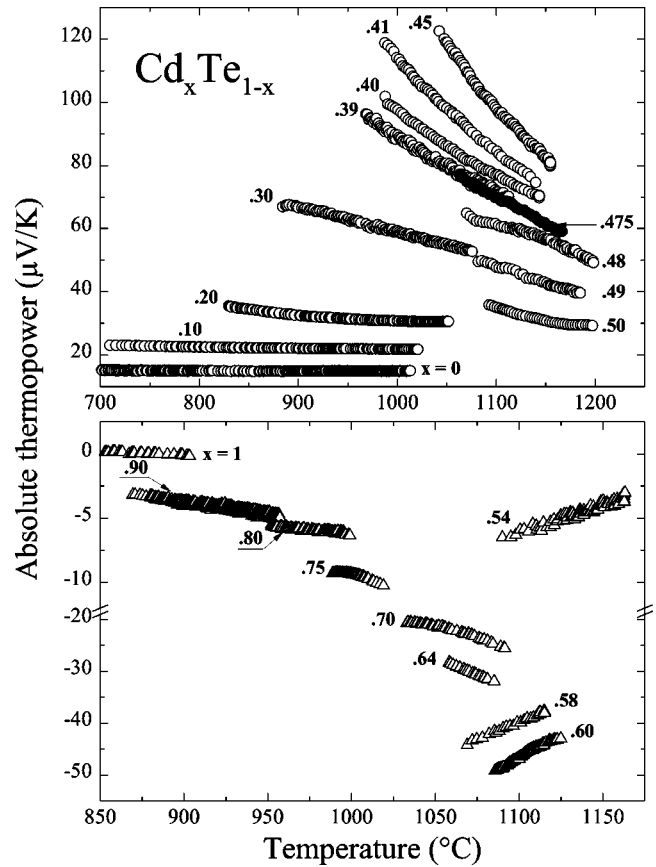


FIG. 2. Absolute thermopower as a function of temperature for liquid $\text{Cd}_x\text{Te}_{1-x}$.

$\rho(d\rho/dT > 0)$. Both alloys have a more metallic behavior with a coordination number (≈ 10) close to that of pure liquid Cd at the melting point.⁴

B. Thermopower

Figure 2 shows the thermopower of liquid $\text{Cd}_x\text{Te}_{1-x}$ as a function of temperature. It is positive between $x = 0$ and 0.5 and shows a negative temperature coefficient ($dS/dT < 0$). The largest value is $120 \mu\text{V/K}$ at 1050°C for $x = 0.45$. For the same composition it has the largest NTC. Near the equiatomic stoichiometry, there is still a ‘‘ p -type’’ behavior. The experimental values are very large if we compare them with liquid metals. The p - n transition can be observed between $0.50 \leq x \leq 0.54$ where S rapidly varies from positive (liquid $\text{Cd}_{0.5}\text{Te}_{0.5}$ has a positive S with a negative slope versus T) to negative (liquid $\text{Cd}_{0.54}\text{Te}_{0.46}$ has a negative S with a positive slope). It stays negative between $x = 0.54$ and 1 . The minimum is reached at $x = 0.60$. The T dependence of S becomes negative for $0.64 \leq x \leq 1$. Within this range, liquid Cd-Te shows a characteristic liquid metals thermopower.

Some isotherms of ρ and S over the entire composition range are shown in Figs. 3(a) and 3(b), respectively, at different T , as well as 30°C above the liquidus. Those of ρ display a sharp peak at a concentration corresponding almost to the equiatomic stoichiometry ($0.50 \leq x \leq 0.54$). The peak may be as high as $10^4 \mu\Omega \text{ cm}$ or more, which is about 100–1000 times higher than ρ of usual liquid metals and metallic alloys. If the composition at which the maximum occurs is

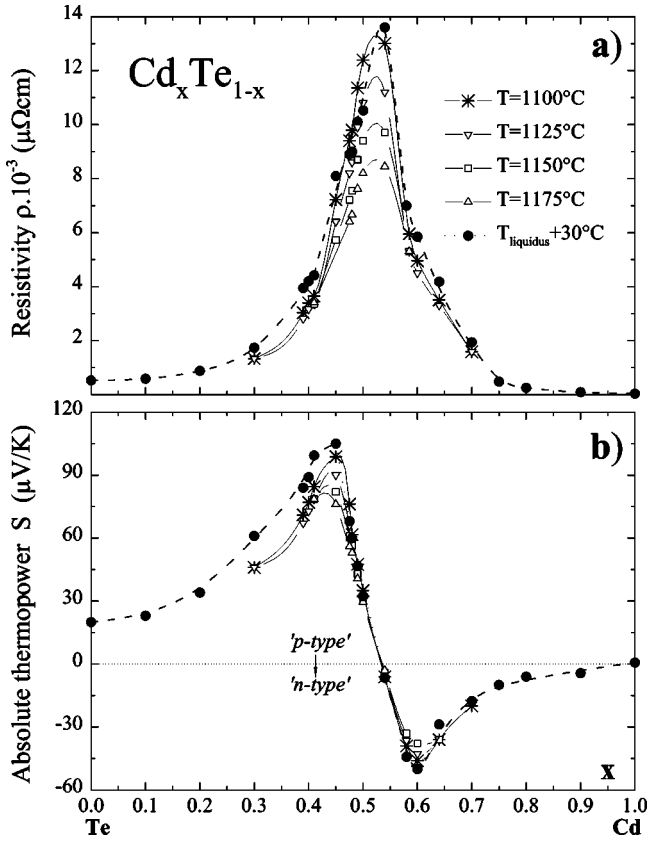


FIG. 3. Electrical resistivity (a) and thermopower (b) as a function of cadmium concentration for liquid $\text{Cd}_x\text{-Te}_{1-x}$. The lines are guides to the eyes.

changed by $\Delta x \approx 0.1$, the resistivity decreases very rapidly by a factor of 3. We observe that ρ reaches its maximum at a composition different from the expected equiatomic one. It can not be excluded that the nominal composition is shifted because of the high vapor pressure of Cd, although the measurements have been repeated giving exactly the same results. As shown in Fig. 3(b), the sign of S changes abruptly near the equiatomic composition, being positive ($S > 0$) at the Te-rich side and negative ($S < 0$) at the Cd-rich side. The x dependence of S exhibits, at 1100°C , a maximum ($S = 99 \mu\text{V/K}$) at $x = 0.45$ and a minimum ($S = -46 \mu\text{V/K}$) at $x = 0.60$. The change in the sign of S , from positive to negative, can be understood in terms of a transition from a hole-like p -type transport to an electron-like one n -type.

IV. DISCUSSION

A. Strong scattering regime ($x \leq 0.39$, $x \geq 0.64$)

In the range $300 \leq \rho \leq 3000 \mu\Omega \text{ cm}$ [$\sim 300 \leq \sigma \leq 3000 (\Omega \text{ cm})^{-1}$], most liquids are characterized by a negative $d\rho/dT$ and can be classified as semimetallic. At the Te-rich side as well as at the Cd-rich side (not too close to pure Cd), the values of σ are located at the border from weak to strong scattering. The conductivity is of the order of $2000 (\Omega \text{ cm})^{-1}$ for pure liquid Te and decreases approaching $x = 0.54$. The density of states (DOS) should deviate to the DOS of a liquid metal (nearly free electrons, NFE). We postulate that a pseudogap,^{8,15} associated with the localization of electrons, appears. In a diffusion regime of the electron mo-

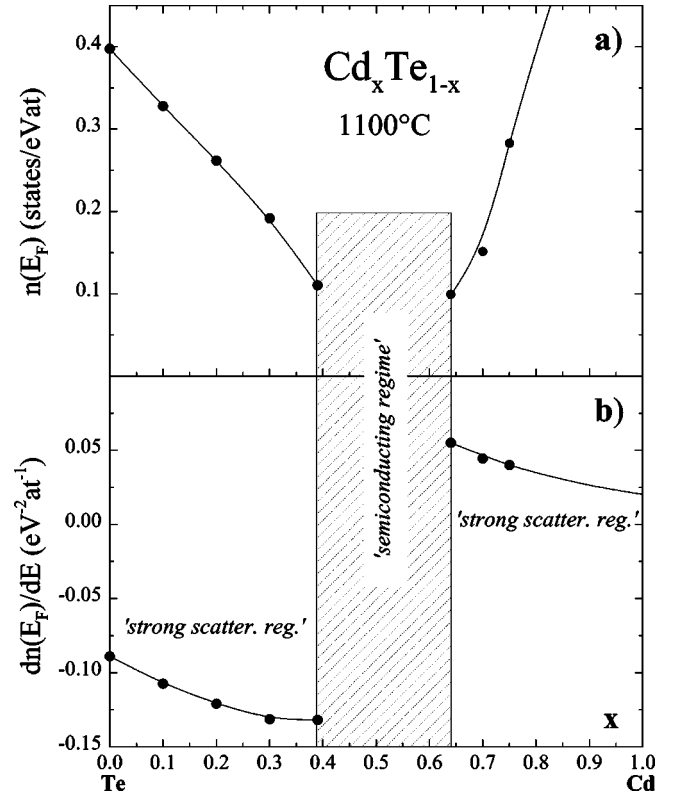


FIG. 4. Density of states (a) and “experimental” $dn(E_F)/dE$ (b) of liquid $\text{Cd}_x\text{-Te}_{1-x}$ at E_F in the framework of the pseudogap approach (strong scattering). Solid circles represent the values estimated from Eqs. (1) and (3). The lines are guides to the eyes.

tion characterized by a magnitude of $300 \leq \sigma \leq 3000 (\Omega \text{ cm})^{-1}$, σ is given by^{8,14}

$$\sigma \cong A \{n(E_F)\}^2, \quad (1)$$

where $n(E_F)$ is the DOS at E_F , the Fermi energy and A is a constant which depends on the interatomic spacing and coordination number as follows:

$$A = 2\pi \frac{e^2 \hbar^3 L}{m^2}. \quad (2)$$

We assume that the mean free path L is comparable with the mean interatomic distance \bar{a} . The value of \bar{a} , for liquid Cd-Te,⁴ is about 2.85 \AA at 1100°C . Then, under the assumption that m in Eq. (2) is equal to the electronic mass, $n(E_F)$ can roughly be estimated from the extrapolation to 1100°C of the experimental value of σ using Eq. (1).

As shown in Fig. 4(a), $n(E_F)$ decreases, from each side, when approaching $x = 0.5$. The decrease of $n(E_F)$ may closely be related to the formation of covalent bonds between Cd and Te ions. In the diffusion regime, S is given by^{8,14}

$$S \cong \frac{-\pi^2}{3|e|} k_B^2 T \left\{ \frac{2}{n(E_F)} \frac{dn(E_F)}{dE} \right\}. \quad (3)$$

Using Eq. (3) and the value of $n(E_F)$, we can estimate $dn(E_F)/dE$ at E_F as shown in Fig. 4(b). The value of $dn(E_F)/dE$ for alloys with $0 \leq x \leq 0.39$ and $0.64 \leq x \leq 1$ decreases gradually with x . The x dependence is fairly different

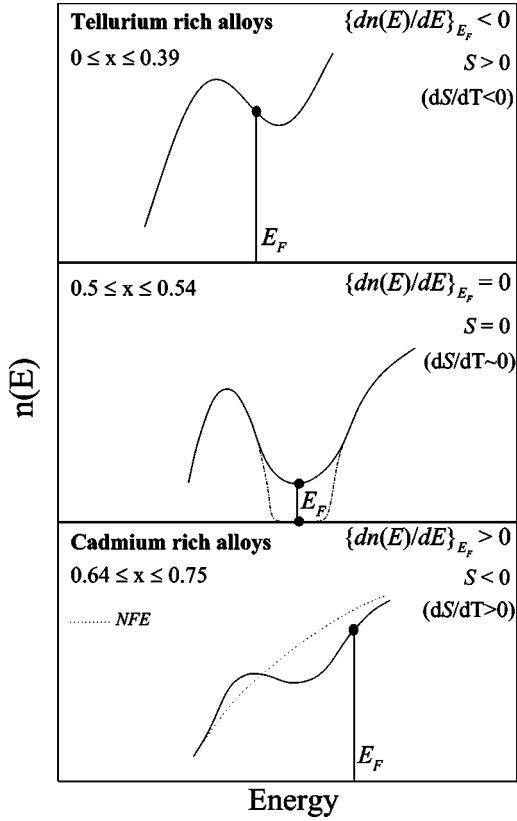


FIG. 5. Schematic representation of the density of states in the pseudogap model (strong scattering regime).

from that observed for the NFE model, suggesting that a deep pseudogap develops in $n(E_F)$ near E_F when approaching $x=0.5$. The origin of the pseudogap may reflect a tendency of both elements to be coordinated (heterocoordination) due to a high electronegativity difference.⁷ The DOS can be described by a pseudogap.^{8,15} As the composition varies from Te to Cd, E_F will move from the left to the right side of the pseudogap as shown in Fig. 5. This explains the sign change in S by a positive value ($\{dn(E)/dE\}_{E_F} < 0$) for Te-rich alloys to a negative one for Cd-rich alloys ($\{dn(E)/dE\}_{E_F} > 0$). The Fermi energy moves through the minimum in the DOS (or zero if there exists a gap). If the conduction is ambipolar with a contribution from both electrons and holes, S can be equal to zero due to $\{dn(E)/dE\}_{E_F} = 0$.

At a qualitative and semiquantitative level, this model can explain in a satisfying manner the observed electronic properties of liquid $\text{Cd}_x\text{Te}_{1-x}$ whose σ is characterized by values in the range of $300 (\Omega \text{ cm})^{-1}$ to $3000 (\Omega \text{ cm})^{-1}$.

Near $x=0.5$, the x dependence of S exhibits the so-called p - n transition. The high ionicity of Cd-Te bonds limits the delocalisation of the electrons and contributes to the semiconducting properties.⁷ Near this composition, we will analyze the electronic properties of liquid Cd-Te by a semiconducting model.

B. Semiconductor regime ($0.39 \leq x \leq 0.64$)

The abrupt decrease in σ on approaching the equiatomic composition indicates a strong tendency for chemical order-

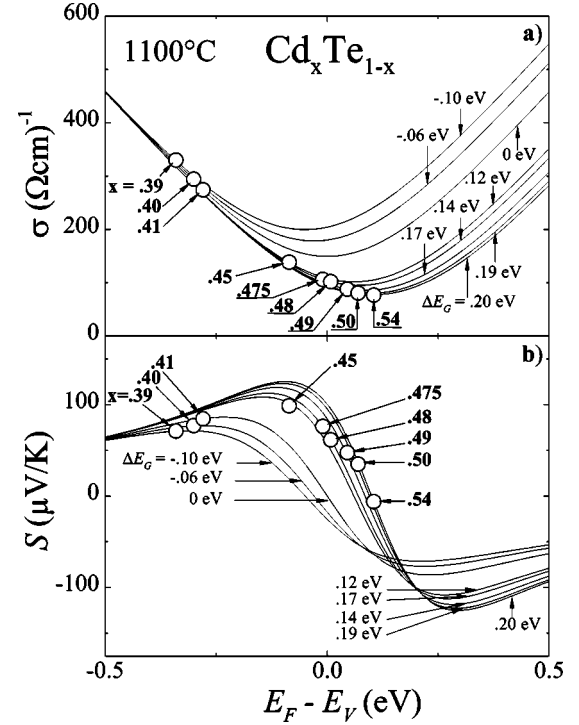


FIG. 6. Variation of σ (curves a) and S (curves b) for liquid $\text{Cd}_x\text{Te}_{1-x}$ with $0.39 \leq x \leq 0.54$ as a function of $E_F - E_V$ for various ΔE_G with $a_c = a_v = 910 (\Omega \text{ cm eV})^{-1}$ within the semiconducting regime. The open circles represent the corresponding experimental values at 1100°C .

ing in this system. The large values of S suggest that a real gap or a very deep pseudogap opens in the DOS. According to experiments and simulation studies^{4,18} of liquid Cd-Te, ΔE_G varies as a function of x as well as T . In this paper therefore we treat the value of ΔE_G as a parameter as described by Okada *et al.*¹⁹

For liquid binary alloys, Enderby and Barnes⁷ have emphasized that the electrical properties of liquid semiconductors can be explained by the equations derived from the Kubo-Greenwood expressions.^{20,21} The electrical conductivity and S are given by

$$\sigma = - \int_{-\infty}^{+\infty} \sigma(E) \frac{\partial f(E)}{\partial E} dE \quad (4)$$

and

$$S = - \frac{k_B}{e} \int_{-\infty}^{+\infty} \frac{\sigma(E)}{\sigma} \frac{[E - E_F]}{k_B T} \frac{\partial f(E)}{\partial E} dE, \text{ respectively,} \quad (5)$$

where $f(E)$ is the Fermi-Dirac distribution function, and $\sigma(E)$ includes all the system-dependent features. Equations (4) and (5) predict that, if E_F crosses the minimum in $\sigma(E)$, a change in the sign of S is likely.

Enderby and Barnes have suggested that the energy dependent conductivity is given, in spirit of scaling theory, by a simple two-band model as follows:

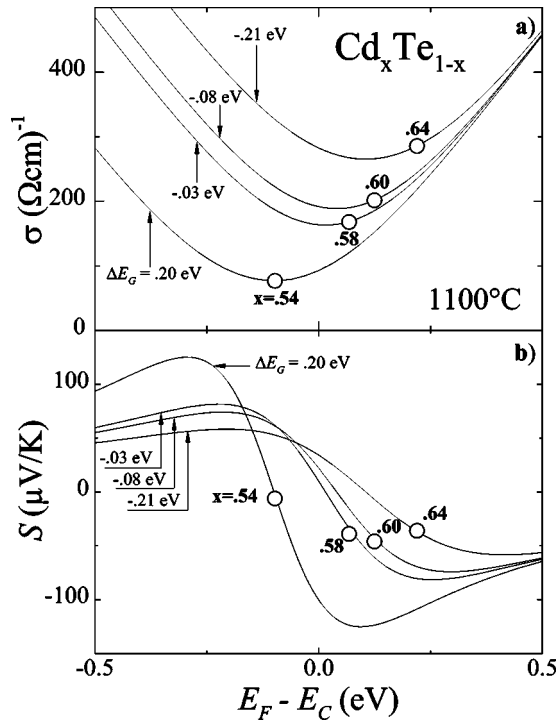


FIG. 7. Variation of σ (curves a) and S (curves b) for liquid $\text{Cd}_x\text{Te}_{1-x}$ with $0.54 \leq x \leq 0.64$ as a function of $E_F - E_C$ for various ΔE_G with $a_c = a_v = 910$ ($\Omega \text{ cm eV}$) $^{-1}$ within the semiconducting regime. The open circles represent the corresponding experimental values at 1100°C .

$$\sigma(E) = \begin{cases} a_c(E - E_C)^v & E \geq E_C \\ a_v(E_V - E)^v & E \leq E_V \\ 0 & E_V \leq E \leq E_C \end{cases}, \quad (6)$$

where a_c and a_v are the coefficients of $\sigma(E)$ for electron and hole states, respectively, and v is a parameter to be determined empirically. We explain our experimental data near the p - n transition with this model.

Putting Eq. (6) into the Kubo-Greenwood formula [Eqs. (4) and (5)], we can estimate, as Enderby and Barnes⁷ for the liquid Mn-Te, the x dependence of σ and S for various values of ΔE_G . For most liquid semiconductors ΔE_G is in the range of 0–0.5 eV. The deep minimum of σ at $x = 0.54$ may be closely related to the large positive value of ΔE_G . For liquid Cd-Te, we have compared our experimental data with Ozols theoretical formula²² using an ambipolar model. For simplicity, the parameters entering Eqs. (4)–(6) are chosen as follows: $v = 1$ (parabolic bands) [if conduction band tailing is ignored $n(E) \propto \sigma^{1/2} \propto E^{1/2}$ at the bottom of the band⁷] and $a_c/a_v = 1$, then the values of a_c and a_v can be estimated from the experimental data of σ at $x = 0.54$. The values obtained are about $a_c = a_v = 910$ ($\Omega \text{ cm eV}$) $^{-1}$ at 1100°C . The position of E_F and the value of ΔE_G can be obtained by adjusting the calculated data of S to the experimental ones.

This allows us to construct $\sigma(E_F - E_V)$ and $S(E_F - E_V)$ (Fig. 6) and $\sigma(E_F - E_C)$ and $S(E_F - E_C)$ (Fig. 7) for different values of ΔE_G . To clarify the results, the origin of the energies is taken at the edge E_V in Te-rich region, as shown in Fig. 6, and at the edge E_C in Cd-rich region, as shown in Fig. 7. The position of $E_F - E_V$ (or $E_F - E_C$) lies on the point

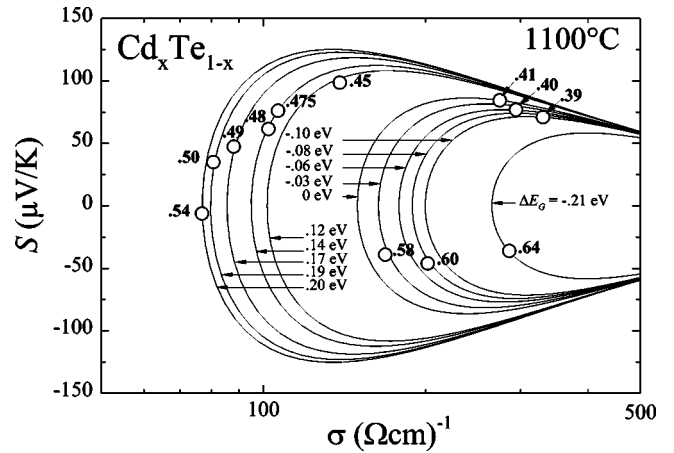


FIG. 8. Plots of S as a function of σ for liquid $\text{Cd}_x\text{Te}_{1-x}$ for $0.39 \leq x \leq 0.64$. The open circles represent the experimental values at 1100°C (semiconducting regime).

where the experimental value of σ is equal to that estimated from Ozols formula²² (open circles).

As shown in Figs. 6(b) and 7(b), we draw the experimental values of S at 1100°C against the position of $E_F - E_V$ (or $E_F - E_C$) which were determined by fitting of σ (open circles). The rapid change in the sign of S from positive to negative reflects the p - n transition as E_F moves across the conductivity gap from E_V to E_C . The data of σ and S suggest that ΔE_G reaches a maximum at a composition near $\text{Cd}_{0.54}\text{Te}_{0.46}$. At this value we obtain $E_F - E_V = 0.1046 \text{ eV}$ and $\Delta E_G = 0.20 \text{ eV}$.

Figure 8 shows the theoretical and experimental correlation between σ and S . There is a cancellation between the positive and negative contributions to S due to electron and hole transport near $x = 0.5$. With the just presented model, we can understand the experimental correlation between S and σ by assuming a small *real* gap $\Delta E_G \cong 0.20 \text{ eV}$ at 1100°C .

In Fig. 9, we plot the conductivity gap $E_C - E_V$ and the position of E_F relative to E_V as a function of concentration between $x = 0.39$ and $x = 0.64$ under the assumption that $a_c = a_v = 910$ ($\Omega \text{ cm eV}$) $^{-1}$. As a result, ΔE_G closes more quickly at the excess Cd side. In Fig. 10, we represent schematically the broadness of ΔE_G and the position of E_F for the same range of x . The formation of covalent bonds, near

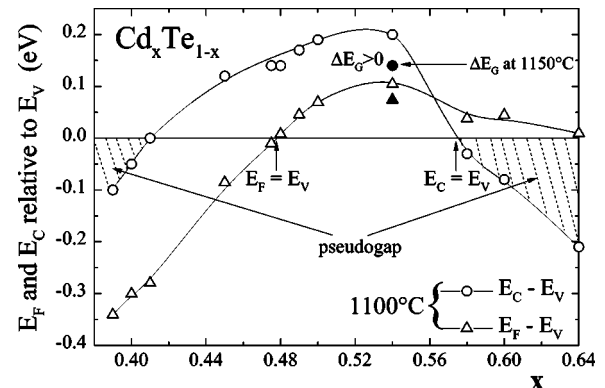


FIG. 9. Broadening of ΔE_G and of the position of the Fermi energy relative to E_V for liquid $\text{Cd}_x\text{Te}_{1-x}$ with $0.39 \leq x \leq 0.64$ at 1100°C and $a_c = a_v = 910$ ($\Omega \text{ cm eV}$) $^{-1}$ (semiconducting regime). The lines are guides to the eyes.

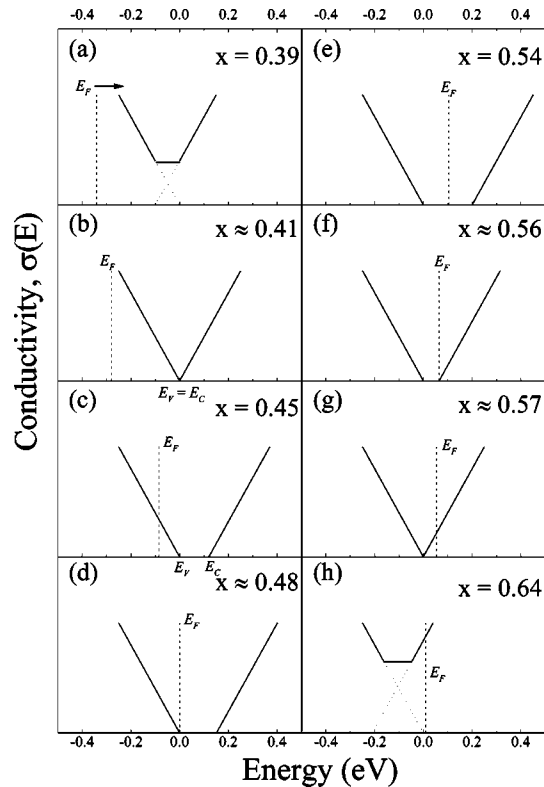


FIG. 10. Schematic representation of $\sigma(E)$ at 1100 °C within the semiconducting regime. The broadness of ΔE_G as well as the position of E_F can be deduced at the different concentrations for liquid $\text{Cd}_x\text{Te}_{1-x}$ following Fig. 9.

$x=0.5$, is well reflected by ΔE_G . The alloys are semiconducting above their melting temperature. The decrease in ρ indicates that ΔE_G closes with T . At 1150 °C and $x=0.54$, we found $\Delta E_G=0.14$ eV and $E_F-E_V=0.1040$ eV as shown in Fig. 9 (solid circle). Godlevsky *et al.*¹⁸ found, by simulation, that heating CdTe much above its melting point leads to substantial structural changes with a transformation to a more close-packed atomic structure, suggesting a gradual semiconductor \rightarrow metal transition with T .

Liquid $\text{Cd}_x\text{Te}_{1-x}$ have negative values of ΔE_G below $x=0.41$ and above $x\approx 0.57$ coming from the overlapping between the conduction and valence bands to form a pseudogap, as indicated by the dotted line in Fig. 10.

V. CONCLUSION

In this work, the electrical resistivity and the thermopower of $\text{Cd}_x\text{-Te}_{1-x}$ were measured in the liquid state, confirming that this system becomes semiconducting around the stoichiometric composition. The composition dependence of S exhibits a p - n transition in the composition range between $0.50\leq x\leq 0.54$. The well-known sigmoid shape for a semiconductor to metal transition was observed.

The sharp peak of the electrical resistivity versus composition observed suggests the existence of a conduction process invoking a conductivity gap near the equiatomic composition and a pseudogap for nearly all other concentrations (except for very rich cadmium alloys and pure Cd). Therefore σ and S were analyzed by assuming an energy-dependent conductivity. According to this analysis the conductivity gap ΔE_G was about 0.2 eV at 1100 °C near the equiatomic stoichiometry. It decreases as a function of Cd and Te content and as a function of increasing T .

Three signatures of a semiconducting behavior near the equiatomic stoichiometry²³ were observed:

- The electrical conductivity is significantly lower than those typical of the metallic state. Its temperature dependence is positive.
- The thermopower changes its sign from p -type to n -type around the equiatomic stoichiometry as the composition varies from tellurium to cadmium.
- The microstructure of liquid Cd-Te shows pronounced local order as evidenced by structural studies.^{4,5}

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