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Thermoelectric power and transverse Nernst-Ettingshausen coefficient of Cd_3As_2 at 300 K

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The thermoelectric power and the transverse Nernst-Ettingshausen coefficient of cadmium arsenide (Cd_3As_2) at room temperature have been calculated as a function of electron concentration, taking into account the HgTe-type inverted electronic-energy-band structure of this material. In the calculation, electron scattering due to polar-optical phonons, charged centers, and acoustical phonons has been considered. Since numerical values were available for every parameter involved except E_d, the acoustical-deformation-potential constant, the latter was treated as an adjustable parameter to fit the theoretical curves with experimental results. It was found that E_d lies between 10 and 20 eV consistent with the approximate value estimated in a previous paper.

I. INTRODUCTION

The electronic-energy-band structure of cadmium arsenide (Cd_3As_2), after having been a question mark for some time, now appears to be similar to that of HgTe.[1] This point of view has been substantiated by several recent publications[2,3] which showed that this model of band structure for Cd_3As_2 is consistent with all the available experimental data. The purpose of the present paper is to calculate the thermoelectric power α and the transverse Nernst-Ettingshausen coefficient P_{NE} of Cd_3As_2 at room temperature as a function of electron concentration, taking explicitly into account the HgTe-type inverted energy-band structure of this material. As in our previous work on the electrical resistivity of Cd_3As_2,[4,5] the calculation of α and P_{NE} is carried out for the mixed mode of polar-optical phonon, charged-center, and acoustical-phonon scattering, using the theoretical results of Zawadzki *et al.*[6-10] In the light of all the results we have recently obtained for Cd_3As_2,[2-5] there is only one unknown in the calculation of α and P_{NE}, which is the acoustical-deformation-potential constant E_d. The latter is used in our procedure as an adjustable parameter to be determined by comparing the calculated results with all the available experimental data on α,[11-23] and P_{NE}.[14,15,20]

II. THEORY

According to Zawadzki and co-workers,[6-10] the general expressions to calculate α and P_{NE} under high statistical degeneracy conditions are given by

$$\alpha = A(R + 5 - 6l) \tag{1}$$

and

$$P_{NE} = A(\mu/c)(R + 2 - 6l), \tag{2}$$

where

$$A = -\frac{1}{3}\left(\frac{\pi}{3}\right)^{2/3} \frac{k_B^2}{\hbar^2} \frac{T}{e} \frac{m^*}{n^{2/3}}, \tag{3}$$

k_B is Boltzmann's constant, c is the speed of light, T is the absolute temperature, $\hbar$ is Planck's constant divided by 2π, e is the magnitude of the electronic charge, and m^* is the electron effective mass at the Fermi energy E_F. R is the effective scattering index which, for the combined scattering mode of polar-optical phonons (op), charged centers (cc), and acoustical phonons (ac), is given by

$$R = \mu\left(\frac{r'_{op}}{\mu_{op}} + \frac{r'_{cc}}{\mu_{cc}} + \frac{r'_{ac}}{\mu_{ac}}\right), \tag{4}$$

where

$$\mu = \left(\frac{1}{\mu_{op}} + \frac{1}{\mu_{cc}} + \frac{1}{\mu_{ac}}\right)^{-1} \tag{5}$$

is the overall electron mobility calculated from the contributions of the three scattering mechanisms considered, namely,[5]

$$\mu_{op} = \frac{\hbar^3 k_F}{2ek_BT(1/\epsilon_\infty - 1/\epsilon_0)(m^*)^2 F_{op}}, \tag{6}$$

$$\mu_{cc} = \frac{\epsilon_0 \hbar^3 k_F^3}{2\pi e^3 (m^*)^2 N F_{cc}}, \tag{7}$$

$$\mu_{ac} = \frac{\pi e \hbar^3 d v_\parallel^2}{k_B T E_d^2 (m^*)^2 k_F F_{ac}}, \tag{8}$$

and

$$r' = r - \frac{k_F}{F_r}\left(\frac{dF_r}{dk}\right)_{k=k_F} \tag{9}$$

denotes the effective scattering index for the electron-scattering mode in question. In Eqs. (6)–(9), ϵ_0 and ϵ_∞ are the low- and high-frequency dielectric constants, k_F is the wave number at the Fermi energy related to the electron concentration in a spherically symmetric degenerate band[24,25] by

$$k_F = (3\pi^2 n)^{1/3}, \qquad (10)$$

N is the total concentration of ionized impurities, d is the material density, $v_\parallel$ is the longitudinal velocity of sound in the material, E_d is the acoustical-deformation-potential constant, and r takes the values 1, −1, and −3 for charged-center scattering, polar-optical phonon scattering, and acoustical phonon scattering, respectively.

The F_r factors for each electron scattering mode are rather involved functions defined in the abovementioned references.[3-10] It is worth mentioning, however, that the mixing of s- and p-like components in the total conduction-band wave function also allows a nonvanishing coupling of electrons to the transverse acoustical phonons, as is transparent in the following expression for F_{ac}:

$$F_{ac} = F^{\parallel}_{ac} + (v_\parallel^2/v_\perp^2)F^{\perp}_{ac}, \qquad (11)$$

where the symbols $\parallel$ and $\perp$ refer to the longitudinal and transverse modes, respectively.

Finally, l is a parameter which is defined as

$$l = \frac{1}{3}\left[1 - \frac{m^*}{\hbar^2}\left(\frac{d^2E}{dk^2}\right)_{k=k_F}\right] = \frac{k_F}{3m^*}\left(\frac{dm^*}{dk}\right)_{k=k_F}, \qquad (12)$$

where $E(k)$ is the dispersion relation for the conduction band. It is clear from Eq. (12) that l gives a measure of the degree of nonparabolicity at the Fermi level of the actual $E(k)$ relation.

III. NUMERICAL RESULTS AND DISCUSSION

In order to calculate α and P_{NE} from Eqs. (1) and (2), we must specify the explicit dependence of m^* and of E_F as a function of electron concentration n (in cm^{-3}) for Cd_3As_2 at room temperature. It has been found that[2,5]

$$\left(\frac{m^*/m_0}{1 - m^*/m_0}\right)^2 = 0.26\times10^{-3} + 1.1\times10^{-15}n^{2/3}, \qquad (13)$$

where m_0 is the free-electron mass, and

$$E_F = 0.095[(1 + 4.4\times10^{-12}n^{2/3})^{1/2} - 1] + 0.00364\times10^{-12}n^{2/3}\ \text{(eV)}, \qquad (14)$$

when measured from the bottom of the conduction band.

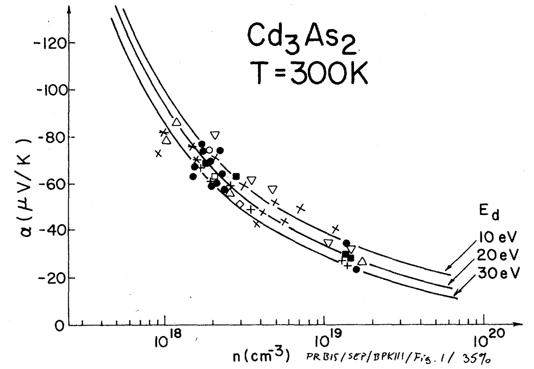

FIG. 1. Thermoelectric power α in Cd_3As_2 at 300 K as a function of electron concentration n. The three curves are calculated for electrons scattered by the mixed mode of polar-optical phonons, charged centers, and acoustical phonons, using three different values of the acoustical-deformation-potential constant $E_d = 10$, 20, and 30 eV. The data points reproduced for comparison are from Refs. 11 (●), 12(◇), 13 (▽), 14 (□), 15 and 19 (△), 16 and 23 (○), 17 and 18 (■), 20 (+), 21 (×), and 22 (*).

The energy-band parameters of Cd_3As_2 at room temperature which are required in the calculation are the $\Gamma_8 - \Gamma_6$ energy gap $E_0 = 0.19$ eV, $E_P = P^2(2m_0/\hbar^2) = 14$ eV, where P is proportional to the $\Gamma_8 - \Gamma_6$ momentum matrix element, and the spin-orbit splitting of the valence band at the Γ point $\Delta = 0.30$ eV.[2,5,26] We also need the numerical values for five material parameters. The latter are d, which is 6.21 g/cm³ for Cd_3As_2,[27] $v_\parallel$, which is 3.5×10^5 cm/sec according to Clavaguera,[28] who attributes this result to Lebourgeois, and according to our own measurements, $v_\perp$, which is reported to be equal to $\frac{1}{2}v_\parallel$ in Cd_3As_2 by various authors,[29-31] and ϵ_0 and ϵ_∞ which are, respectively, 36 and 12 according to previous fits with experimental transport data.[4,5] Assuming that there is only one species of donor impurity and that it is singly ionized, and ignoring compensation effects [i.e., $N = n$ in Eq. (7)], the only parameter which

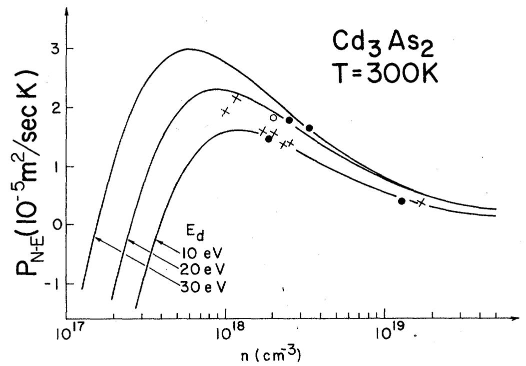

FIG. 2. Transverse Nernst-Ettingshausen coefficient P_{NE} in Cd_3As_2 at 300 K as a function of electron concentration n. The three curves are calculated for the mixed mode of polar-optical phonon, charged-center, and acoustical phonon scattering, using three different values of the acoustical-deformation-potential constant $E_d = 10$, 20, and 30 eV. Experimental data are from Refs. 14 (○), 15 (×), and 20 (●).

TABLE I. Calculated coefficients at $T = 300$ K for different electron concentrations, assuming $E_d = 10$ eV.

Electron concentration (cm^{-3})	1×10^{17}	5×10^{17}	1×10^{18}	5×10^{18}	1×10^{19}
E_F (eV)	0.0387	0.0927	0.131	0.272	0.365
m^*/m_0	0.0220	0.0304	0.0362	0.0567	0.0695
μ_{cc} ($\mathrm{m}^2/\mathrm{V\,sec}$)	36.8	17.8	12.3	4.75	3.10
μ_{ac} ($\mathrm{m}^2/\mathrm{V\,sec}$)	97.3	47.7	33.9	13.6	8.66
μ_{op} ($\mathrm{m}^2/\mathrm{V\,sec}$)	2.21	2.31	2.14	1.52	1.26
μ ($\mathrm{m}^2/\mathrm{V\,sec}$)	2.04	1.96	1.73	1.06	0.812
r'_{cc}	0.809	0.881	0.895	0.914	0.920
r'_{ac}	−2.30	−2.00	−1.97	−2.13	−2.27
r'_{op}	−0.712	−0.779	−0.870	−1.03	−1.06
l	0.158	0.237	0.262	0.292	0.296
α (μV/K)	−349	−142	−100	−50.1	−38.7
P_{NE}(10^{-5} $\mathrm{m}^2/\mathrm{sec\,K}$)	−8.17	0.669	1.56	0.896	0.524

remains to be specified is E_d. A rough estimate of this parameter which is $E_d \sim 10$ eV comes from our previous electronic-energy-band-structure calculations.[3] This value of E_d was subsequently used in all our electron-mobility calculations in $\mathrm{Cd_3As_2}$,[5] as well as in the $\mathrm{Cd_{3-x}Zn_xAs_2}$ alloy system ($x \lesssim 1.5$).[32] In the following, we treat E_d as an adjustable parameter to be determined by comparing the calculated results of α and P_{NE} with all the available experimental data. This procedure has been used successfully by Zawadzki and coworkers[6,8,10] for the determination of the deformation-potential constant of InSb. Figure 1 illustrates the 300-K thermoelectric-power results obtained by various authors[11-23] on a number of $\mathrm{Cd_3As_2}$ samples, as well as theoretical curves computed from the equations of Sec. II for three different values of E_d, namely, 10, 20, and 30 eV, respectively. The same is done in Fig. 2 for the transverse Nernst-Ettingshausen coefficient of $\mathrm{Cd_3As_2}$ at 300 K. Table I summarizes a few of the results leading up to the $E_d = 10$ eV curves for α and P_{NE}. It is clear from Fig. 1 that the predicted behavior of α with n is consistent with the experimental results. The small spacing between the curves illustrates the weak dependence of the thermoelectric power on acoustical phonon scattering. This insensitivity is due to the factor 5 which appears in the expression $R + 5 - 6l$ of Eq. (1) and which dominates small possible changes of R resulting from different relative importances of the various scattering modes. Taking also into account the scatter of the data, it can only be stated that E_d lies between 10 and 30 eV.

On the other hand, Eq. (2) contains the factor $(R + 2 - 6l)$. Since for high electron energies in a nonparabolic band such as the conduction band of $\mathrm{Cd_3As_2}$ $2 - 6l \sim 0$ (see, for example, Table I), the Nernst-Ettingshausen coefficient is almost directly proportional to the scattering index R. This explains the larger spacing between the curves of Fig. 2 which even predict a change in sign of the Nernst-Ettingshausen coefficient at sufficiently low carrier densities. With the available data, it may reasonably be stated that E_d lies between 10 and 20 eV. A much more precise value of E_d could be obtained from samples with $n < 10^{18}$ cm^{-3} since then the Nernst-Ettingshausen coefficient would approach zero (see Fig. 2). It now appears that such samples are available.[33]

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