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Comments and Addenda

The section Comments and Addenda is for short communications which are not appropriate for regular articles. It includes only the following types of communications: (1) Comments on papers previously published in **The Physical Review** *or* **Physical Review Letters**. *(2) Addenda to papers previously published in* **The Physical Review** *or* **Physical Review Letters**, *in which the additional information can be presented without the need for writing a complete article. Manuscripts intended for this section must be accompanied by a brief abstract for information-retrieval purposes. Accepted manuscripts follow the same publication schedule as articles in this journal, and page proofs are sent to authors.*

Evidence for the existence of an Einstein-type phonon mode in the resistivity of pure cadmium

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The temperature-dependent resistance of pure cadmium crystals, parallel and perpendicular to the hexagonal axis, can be described as the sum of three contributions: a term $\propto T^3$, a Grüneisen-Bloch term, and an exponential term dominant for $T > 5$ K. As suggested earlier by Hsu and Falicov, this term can be interpreted as an extra scattering contribution due to an Einstein-type phonon mode with $40 < \Theta_E < 50$ K.

In order to explain an observed[1] exponential temperature dependence of the ratio between the Hall resistivity and the zero-field resistivity in cadmium in the temperature range 10–40 K, Hsu and Falicov[2] (hereafter referred to as HF) proposed that this behavior is caused by an Einstein-type phonon mode with a characteristic temperature Θ_E. This mode is the result of a rather complicated weighing of the combination of TA and TO modes of almost equal frequency that are constant on a large range in the Γ K direction. Using a simple, workable model of the Fermi surface, HF succeeded in a semiquantitative description of the observed relations after the derivation of electron (τ_e) and hole (τ_h) relaxation times. Though their computation required the adjustment of several parameters, it described the maximum in the Hall resistivity that was observed around 10 K, and also indicated that $42 < \Theta_E < 48$ K, in agreement with the experimental best-fitting value of 43 K.

An effect of this phonon mode on the resistivity is to be expected, but the data that were available at that time were not precise enough to attempt a quantitative analysis.

New resistivity data[3] (ρ parallel to the c axis) that were obtained with two pure crystals have been combined for the analysis. The resistance of crystal Cd-1 ($r_{1.4\,\mathrm{K}} = 7.9\times 10^4$), r_T is the resistance ratio between 273 K and T was observed to vary as $\propto T^3$ for $T < 3$ K. This was also observed for a much purer, zone-refined crystal Cd-2 ($r_{1.4\,\mathrm{K}} = 15.2\times 10^4$) with the difference that a change of slope was observed around $T \approx 1.3$ K. This affects the error in the determination of the residual resistance at 0 K for this crystal. Since for crystals of this purity the residual-resistance value is negligible with respect to the temperature-dependent part of the resistance above, say 5 K, the data for Cd-1 for $1.2 \leq T \leq 9$ K were combined with those for Cd-2 for $10 \leq T \leq 25$ K.

The high precision in the data below 10 K are obtained by an experimental method that has been described before.[4] Below 9.5 K the precision in the resistance values is better than 0.01%, for higher temperatures it is 0.2%. Temperatures were read within 0.2%, so that the quantity of interest, ρ/T^5, is precise within 1.2% over the temperature range in consideration. The error in the absolute value of ρ is determined by the choice of the ice-point resistivity $\rho_{\parallel} = 7.73$ $\mu\Omega$ cm $\rho_{\perp} = 6.35$ $\mu\Omega$ cm, taken from the literature.[5]

The temperature-dependent part of the resistivity is plotted in Fig. 1. A maximum at $T \approx 9.5$ K is preceded by a low-temperature branch varying as T^{-2}, which corresponds to $\rho \propto T^3$. Similar data have been plotted in Fig. 2 for a previously measured crystal[6] with ρ perpendicular to the c axis ($\rho_{\perp}$). For this crystal, $r_{4\,\mathrm{K}} = 1.75 \times 10^4$ and the measurements, in particular those of the temperature, were less precise. Apart from the spread in the data, the behavior to $\rho_{\perp}$ is similar to that of $\rho_{\parallel}$. It is appropriate[7] to analyze the $\rho(T)$ relation in the two-band approximation:

$$\rho \propto \frac{m_e}{\tau_e}\frac{m_h}{\tau_h} \Big/ \left(\frac{m_e}{\tau_e} + \frac{m_h}{\tau_h}\right) . \tag{1}$$

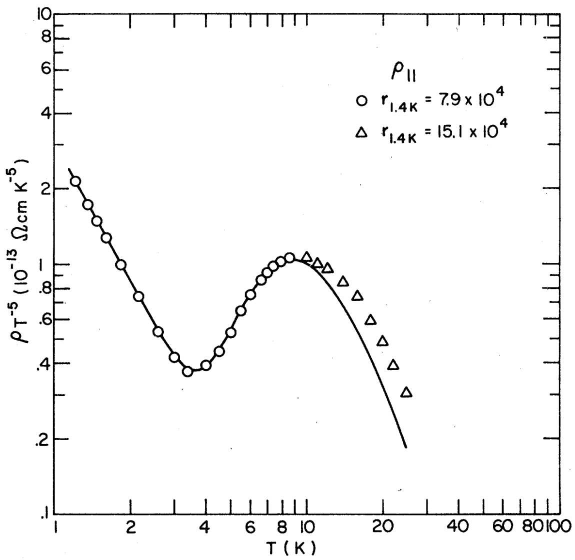

FIG. 1. Temperature-dependent part of the resistivity for two crystals grown parallel to the hexagonal axis plotted as ρT^{-5} vs T. The drawn curve represents the result of a numerical analysis, in which the scattering by an Einstein-type phonon mode is incorporated.

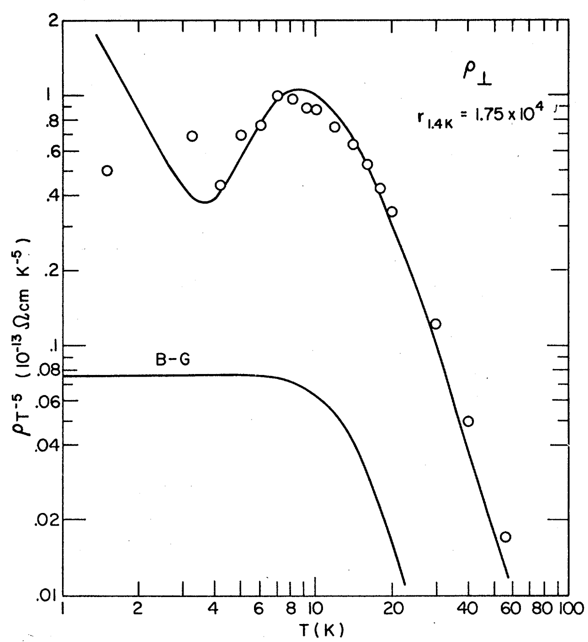

FIG. 2. As Fig. 1, but for a crystal grown perpendicular to the hexagonal axis (Ref. 6). The drawn curve is the same as in Fig. 1. The curve marked B-G represents the contribution by the Bloch-Grüneisen term.

Expressions for τ_e^{-1} and τ_h^{-1} are given in HF, and they consist of an impurity term, a Bloch-Grüneisen term, and the term for the newly introduced scattering, $\propto[\exp(\Theta_E/T)-1]^{-1}$. In the following we will omit for the pure crystals the temperature-independent impurity term. Furthermore, in order to account for the observed maximum in the Hall coefficient (HF) introduced a nonisotropic contribution by the Einstein mode, namely, in τ_h^{-1} only. Substituting HF's expressions for τ_e^{-1} and τ_h^{-h} in (1), one can then attempt to find from the measured data values for the coefficients in the formal expressions that represent the three different scattering processes.

Some simplifications present themselves. For any $\Theta_E > 30$ K, the contribution of the exponential is negligible for temperatures below 3 K. Also, below $T = 5$ K, the integrals $J_5(\Theta/T)$ which determine the Bloch-Grüneisen contribution decrease only of the order of 3% towards lower temperatures.[7] In the two-band approximation the Bloch-Grüneisen (BG) term can be written

$$\rho_{BG}/T^5 = \frac{\alpha_2[J_5(\Theta_e/T)]\,[pJ_5(\Theta_h/T)]}{J_5(\Theta_e/T)+pJ_5(\Theta_h/T)} \equiv F(T) \qquad (2)$$

in which (HF) $p = (k_{Fe}a/h_{Fh}a)^3(m_h/m_e)^2 = 0.189$, wherein the ratio between electron and hole surface dimensions equal 2 and $m_h/m_e = \frac{1}{6.5}$, $\Theta_e = 105$ K, and $\Theta_h = 77.6$ K.

The plot of ρT^{-5} vs T^{-2} results for $T \leqslant 3$ K in a straight line $\alpha_1 T^{-2} + \alpha_2$ $(F(T)$, with $\alpha_1 = 3.08 \times 10^{-13}\ \Omega\,\mathrm{cm\,K^{-3}}$ and $\alpha_2 = 3.89 \times 10^{-16}\ \Omega\,\mathrm{cm\,K^{-5}}$. This indicates additative scattering. It is interesting to note that adding a term proportional to T^3 in either τ_e^{-1} or τ_h^{-1} in (1) does not yield positive values for α_1. Addition to both τ^{-1} expressions and weighing the hole contribution with the factor p (for which there is no argument) yields, within 1% or 2%, the same values for α_1 and α_2. This is then a consequence of the practically constant values of the integrals J_5.

In the case of the exponential term the factor p can be absorbed in its unknown coefficient α_3 when, as suggested by HF, this term should only be added into τ_h^{-1}. In the temperature region where this term dominates, the contribution by the term proportional to T^{-2} is small, but not negligible compared to the BG term. However, neither an asymmetric nor a symmetric addition of the exponential term in τ_e^{-1} and τ_h^{-1} in (1) leads to real or consistently positive values for this contribution. A "series" addition is then the simplest to consider even though no physical or mathematical argument can be given for this approach. The difference between the measured data ρT^{-5} and the sum of (2) and $\alpha_1 T^{-2}$ should then yield values for $\alpha_3 T^{-5}[\exp(\Theta_E/T)-1]^{-1}$. Below

≈ 10 K this corresponds to $\alpha_3 T^{-5}\exp(-\Theta_E/T)$ and one finds $\Theta_E = 44.27$ K with $\alpha_3 = 7.50\times 10^{-7}\Omega\,\mathrm{cm}\,\mathrm{K}^{-5}$. This result was obtained for $3.4 \leq T \leq 8$ K (in which range $\alpha_3[\exp(\Theta_E/T) - 1]^{-1}$ increases by a factor 2.2×10^3) for 11 data points and a correlation factor of 0.999 94.

Notwithstanding the spread of the data for the case $\rho_\perp$, the rise up to the maximum and the location of the maximum are in agreement with the measured data. At higher temperatures the analysis underestimates the resistivity parallel to the c axis. On the other hand, the agreement at higher temperatures is astonishingly good in the case of $\rho_\perp$. It is known[8,9] that the anisotropy in the electrical resistivity ($\rho_\parallel/\rho_\perp$) and the thermal resistivity ($w_\parallel/w_\perp$) decreases drastically below $T = 23$ K and $T = 10$ K for the electrical and thermal cases, respectively. These results, combined with those of the present analysis, lead to the suspicion that the large anisotropy in these transport quantities is caused by an extra, as yet unaccounted for scattering for transport parallel to the c axis above, say, 10 K.

Though we have demonstrated without any physical argument that a contribution of scattering by an Einstein-type phonon mode exists with a Θ_E in agreement with HF results $42 < \Theta_E < 48$ K and in agreement with the previously reported result (42.9 K) on the ratio R/ρ, the analysis is by no means conclusive. In particular there is the discrepancy that it was impossible to find this contribution numerically through its incorporation in τ_h^{-1} as required in the analysis by HF.

It should be noted that the eventual existence of this mode is a property of the Cd and not due as reported recently, to the addition of impurities having less mass.[10] The observation is to be considered as another example[11] of the effect of umklapp processes by a particular phonon mode at low temperatures. The occurrence of the term proportional to T^3, which seems to be impurity dependent, is difficult to explain without more data.

The behavior below 1.3 K for very pure material has to be investigated experimentally, also more precise data for $\rho_\perp$ are needed. The unfortunate circumstance that further investigation by the author is not possible was a justification for the publication of these, as yet incomplete results.

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