Generalized phonon-connected relations in the Hall resistivity of cadmium*

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(Received 4 June 1975)

An analysis of Hall resistivity data for Cd and Cd-alloy crystals previously published by Lilly and Gerritsen has led to the following, rather unexpected results. The coefficient (A_1) of the term in first order in H, the magnetic field parallel to the hexagonal axis, varies as $A_1 \propto \rho_{11}^{-1} e^{-43/T}$ in the temperature range $8 \leq T \leq 200$ K (ρ_{11} is the resistivity parallel to $\langle 11\bar{2}0 \rangle$). The difference between the measured Hall resistivity and $A_1 H$ passes zero for all samples at $T \approx 11$ K. This temperature can be connected to the onset of strong, specific phonon modes. A further relation, that may have less physical significance is that in the temperature range $10 < T < 20$ K, where the Hall resistivity passes through a maximum, the field H_m at the maximum varies as $H_m \propto \rho_m^{-1} e^{-109/T_m}$. The temperature 109 K compares to the temperature $\Theta_R = 112$ K that has to be used in the Bloch-Grüneisen relation to describe $\rho_{11}(T)$ for temperatures above ≈ 8 K.

I. INTRODUCTION

During the last few years results on the measurement of the Hall resistivity (ρ_{21}) in cadmium crystals of different purity have been published[1,2] and the general behavior of $\rho_{21}(H, T)$ has been confirmed by different authors.[3,4] A striking effect is the sign reversal that occurs when using crystals of sufficiently high purity at temperatures below 6 K. The main purpose of Refs. 1 and 2 was to describe the dependence of this effect on temperature, magnetic field, and impurity concentration. The results led to an interpretation[5,6] based on the transfer of hole orbits into electron orbits on branches of the Fermi surface in the basal plane of the crystal (intersheet scattering). Some aspects of the temperature dependence of the Hall resistivity, which were visible in the results, were intentionally not discussed in Ref. 2 and will be considered in detail in the present paper. These aspects are: the deviation from linearity in the observed $\rho_{21}(H)$, curves at constant T and the maximum at $T \approx 16$ K that is a persistent feature in the $\rho_{21}(T)$ curves at constant H.

In a recent paper[7] it was shown that when samples with certain amounts of doping and which exhibit a positive residual Hall resistivity are used, the contribution to $\rho_{21}(H)$ by higher-order terms in H is negative below 8 K. This suggests that in those cases intersheet scattering still plays a role. A qualitative discussion of the 16-K maximum[8] led to the conclusion that this maximum, because it appeared to be observed with varying magnitude for both pure and impure crystals, is the result of phonon effects rather than transitions from low-magnetic-field ($\omega\tau < 1$) regimes in different orbits. Both interpretations have appeared in the literature.[9]

These observations indicate that a simple relation between the maximum in ρ_{21} (further indicated as ρ_{21m}) and $\omega\tau$ is very improbable, because the temperature at which ρ_{21m} occurs lowers when the magnetic fields are decreased and increases when the amount of impurities is increased. Indeed, it has been shown (Ref. 2, Fig. 4) that for $T > 10$ K a rough presentation of ρ_{21} is possible in some kind of Kohler diagram, where $\rho_{21}(H, T)/\rho(0, T)$ is plotted against $rH \propto (\omega\tau)$ and r is the ratio between the resistance (or resistivity ρ_{11}) at 273 K and at T. (The zero-field resistivity ρ_{11} will simply be indicated by ρ throughout the paper.) For temperatures below 25 K the data may deviate as much as by a factor of 2 from a curve that was drawn through the data for a pure crystal. This representation fails drastically at the onset of the intersheet scattering, which in many cases is preceded by a large positive peak in $\rho_{21}(T)$. The general behavior of $\rho_{21}(T)$ is illusrated in Fig. 1, taken from Ref. 2. A further analysis of the available data, which was only partly given in Ref. 2, could result in the construction of general relations between the quantities involved. It will be shown that such relations do exist, that they support an explanation based on phonon scattering for ρ_{21m}, and that they may present a clue to the existence of rather specific phonon modes.

The experimental details have been discussed in Refs. 2 and 8. For the present it is of interest to note that all samples were single crystals oriented such that the magnetic field was parallel to $\langle 0001 \rangle$ and the dc current parallel to $\langle 11\bar{2}0 \rangle$. The reason for choosing these particular alloys and their composition (not more than a few hundred ppm silver or indium) has been given in Ref. 2. Most of the Hall voltages were obtained in the temperature range 1.4–80 K by current and field reversal. In some cases a few higher temperatures were utilized. The following will be dis-

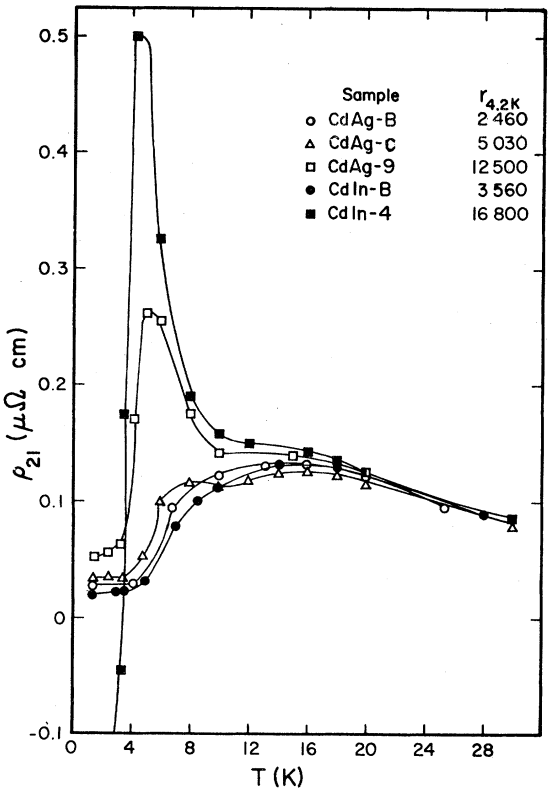

FIG. 1. Hall resistivity for a few samples. Observable are the sign reversal (for CdIn-4), the large peak for two samples, and the maximum at 16 K. This figure has been taken from Ref. 2 (Fig. 2).

cussed successively: $\rho(T)$; $\rho_{21}(H)$ for constant T; and relations pertaining to ρ_{21m} for $10\ \mathrm{K} \leq T \leq 20\ \mathrm{K}$.

II. ZERO-FIELD RESISTIVITY

As might be expected from simple arguments, the resistance ratio $r(T) = R(273)/R(T) = \rho(273)/\rho(T)$ is an important parameter in the relations that will be given. This, in a sense, should be obvious because $\rho(T)$ indicates the temperature dependence of the electron-phonon and impurity scattering. For not too low temperatures, for a pure metal, $\rho(T)$ is described[10] fairly well by the Grüneisen-Bloch relation

$$\rho \propto T^5 J_5(\Theta_R/T) \ . \qquad (1)$$

In this expression J_5 is a well-known integral that accounts for the scattering of the conduction electrons by normal phonon processes when the phonon umklapp processes are excluded. The characteristic temperature Θ_R follows from the matching of Eq. (1) to the experimental data and is, in general, not equal[11] to the Debye temperature Θ_D which, because the derivation of Eq. (1) is based on the Debye phonon model, appears formally in Eq. (1). Furthermore Eq. (1) should hold for a free-electron spherical Fermi surface.

An approximate experimental value for Θ_R follows from the ratio of two resistance values of the same sample taken in the low-temperature regime ($J_5 = \text{const}$) and for a $T > \Theta_D$, where $J_5 \propto (\Theta_D/T)^4$. For Cd, $\Theta_D = 210.3 \pm 2$ K,[12] and from this estimate, it follows that $\Theta_R \simeq 110$ K. Indeed, fitting Eq. (1) for $1/r(T) \propto \rho(T)$ to the r value at Θ_R from the observed $r(T)$ curves, a very reasonable fit of Eq. (1) was obtained for $\Theta_R = 112$ K. This is illustrated in Fig. 2 ($T < 150$ K) for two alloys of different purity and in Fig. 3 for a few more samples in the temperature region of interest ($10 \leq T \leq 20$ K). In this temperature range, where J_5 for $\Theta_R \approx 100$ K varies[13] by a factor of 2.5, differences of a few degrees in the choice of Θ_R affect the fit with the experimental data in an observable manner. However, it is obvious that $100 \leq \Theta_R \leq 112$ K. Deviations larger than those that depend on the choice of Θ_R, do occur when the impurity scattering is dominant. This can be seen for the low-resistance Cd samples (when $T \approx 10$ K) and for two high-resistance alloys. Furthermore, as demonstrated by the data for the sample CdIn-3, a correction for the residual resistance seems to imply that the magnitude of the impurity resis-

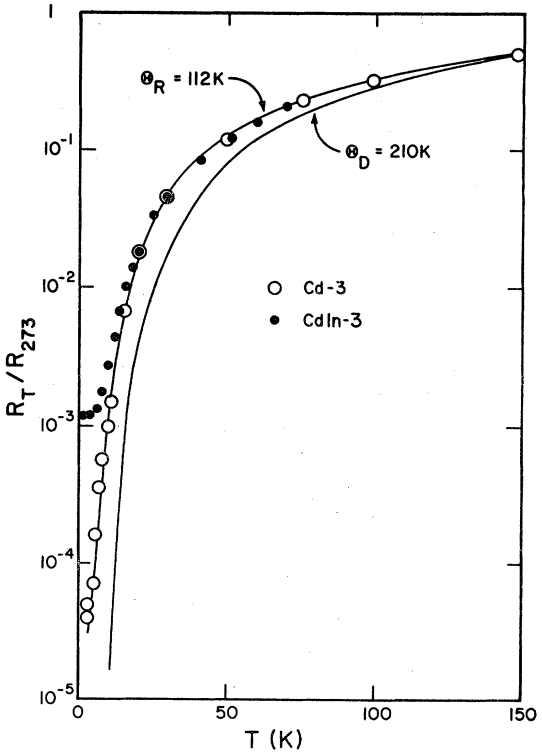

FIG. 2. Reduced resistivity parallel to $\langle 11\bar{2}0 \rangle$ for two samples. Drawn curves represent the Grüneisen-Bloch relations for $\Theta_R = \Theta_D = 210$ K and $\Theta_R = 112$ K.

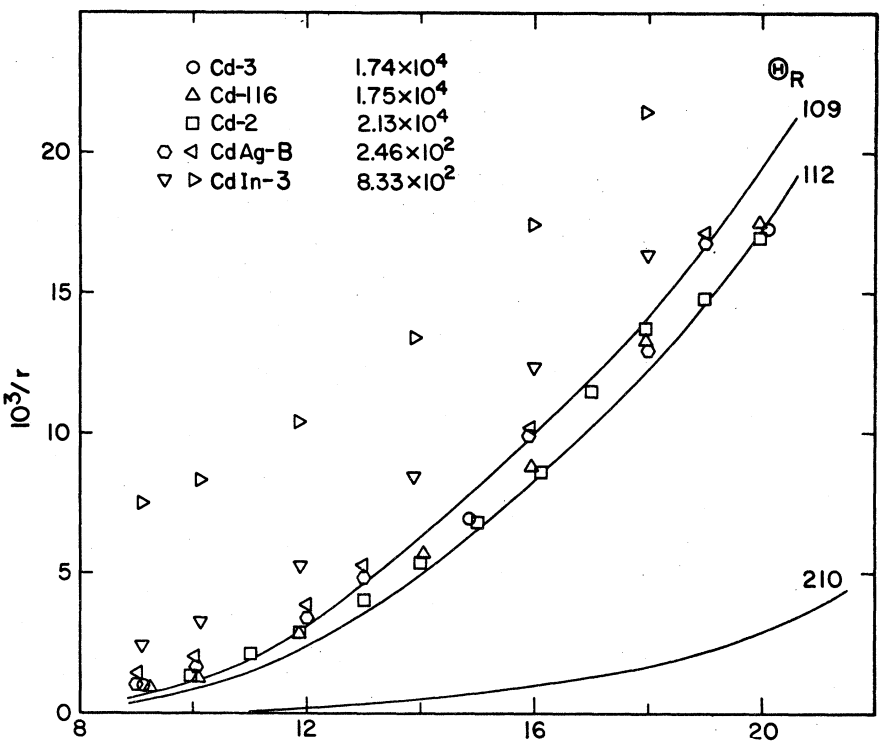

FIG. 3. Reduced resistivity for $9 \leq T \leq 20$ K. In this and the following figures the value for r at 4.2 K is listed after the sample index. Curves represent the Grüneisen-Bloch relation for three different values for Θ_R. The measured $1/r$ values for the last two impure samples have been plotted after subtraction of their residual resistance ratio (indicated by a hexagon and inverted triangle).

tance has its influence on Θ_R as well; a value $\Theta_R < 109$ K would be more satisfactory. Deviations of the Matthiessen rule are known to affect the temperature dependence of the resistivity[14]; in the present case these effects are small. They do occur in only a few samples and they will not be discussed further. Considering the restrictions under which Eq. (1) has been derived, it is not expected that Eq. (1) be applicable for the case of cadmium. In a direction perpendicular to $\langle 0001 \rangle$ no continuous Fermi surface exists, and one would expect a larger influence of umklapp processes. The fact that $\rho(T)$ follows Eq. (1) in first order almost suggests that normal scattering processes with particular phonons that are characterized by a characteristic temperature different from Θ_D have a dominant effect.

Below $T \approx 10$ K, $\rho \propto T^5$ for $\Theta_R = 210$ K, whereas for $\Theta_R = 112$ K, the onset of the T^5 region would occur at $T \approx 8.5$ K. The present data indicate that deviations from the T^5 law do occur below 6 K; the true temperature dependence is presently under investigation.

III. THE DEPENDENCE OF ρ_{21} ON THE FIELD

During the measurements it was found that field reversal did not indicate the presence of a contribution $\rho_{21} \propto H^2$ within a limit less than the relative uncertainty[2] of 5% for the smallest voltages ($\sim 10^{-8}$ V) measured. When the data were corrected for the voltage contribution, which arose because the two Hall probes were not exactly in the sample plane, the magnitude of the residue voltage, its independence on the magnetic field, and its sample by sample variation suggested that it was due to small thermoelectric voltages that are usually present in experiments of this kind. The $\rho_{21}(H, T)$ curves were, therefore, analyzed as

$$\rho_{21}(H, T) = A_1(T) H + A_3(T) H^3 + A_5(T) H^5 . \quad (2)$$

Since not more than six (and often not more than five) field values were measured at a given temperature, no significance can be attributed to the individual values of the coefficients A_3 and A_5 for each individual sample, but the temperature dependence of A_1 and the quantity $\Delta\rho_{21} = \rho_{21} - A_1 H$ for all samples taken together will be of interest. In all cases a description of ρ_{21} by Eq. (2) was satisfactory within less than 5% in ρ_{21}. In some cases the Hall voltages for the lowest fields (1.8 kG and 4.5 kG) were below the limit of measurement (10^{-8} V) and were omitted. In Figs. 4(a) and 4(b), $A_1(T)$ and $\Delta\rho_{21}(T)/\rho_{21}(T)$ are plotted for samples of different purity. In each category only a few samples are represented. Samples with $r_{4.2}$ values between the given ones can be represented by curves between those that have been drawn. For all samples, A_1 passes through a maximum which is, in general, close to the temperature where ρ_{21m} is observed.

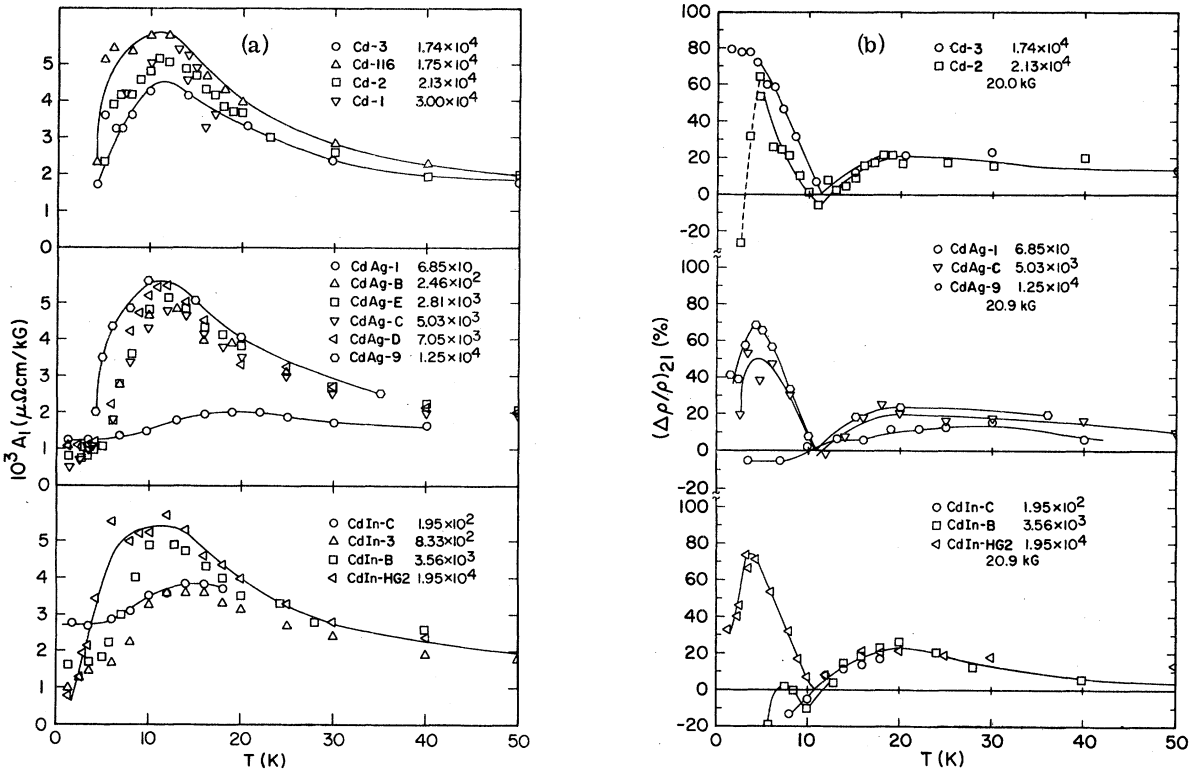

FIG. 4. (a) Temperature dependence of the coefficient of the linear term in $\rho_{21}(H)$ for some samples in each group. For the sake of clarity the data have been connected by a curve in each group for two samples only. (b) Percentage difference between ρ_{21} and $A_1 H$ against temperature for given fields for the samples in Fig. 4(a). Note that the difference is zero for $T \approx 11$ K.

The curves for $\Delta\rho_{21}/\rho_{21}(T)$ are characterized by a rather sharp decline to $\Delta\rho_{21} = 0$ at a temperature that is approximately 11 K and does not seem to change markedly for a variation in purity. This is illustrated clearly [Fig. 4 (b)] by the $\Delta\rho_{21}/\rho_{21}(T)$ curve for the high-residual-resistivity sample CdAg-1, for which the maximum for A_1 and ρ_{21m} have been shifted to around 20 K. In a few cases $\Delta\rho_{21}$ continues through zero to a negative value that is larger than 5% maximum error, which, when the $\Delta\rho_{21} < 0$ values for $T < 5$ K were included, led to the reasoning[7] concerned with the intersheet-scattering phenomena in these alloys. For temperatures above the maximum of A_1 the curves in Fig. 4(a) suggest that $A_1 \propto T^{-1}$. When plotted in this manner, the different samples were represented by individual curves with different slopes in the high-temperature range. It was assumed that a better representation could be achieved when $A_1(T)$ could be connected to the scattering parameter $\tau(T)$. Should a strong correlation between these parameters exist, one could hope that a plot of A_1/r, which is equivalent to A_1/τ, against temperature would describe all data and provide pertinent information about the correlation between A_1 and τ. This turned out to be the case. Plots of A_1/r against $1/T$ are given for the three different groups of samples in Fig. 5. The straight line is the result of a rms analysis for the pure Cd data

$$A_1/r = 5.40\times10^{-4} e^{-42.9/T}\ \mu\ \Omega\ \text{cm/kG}, \qquad (3)$$

with a correlation factor equal to 0.998. Marks at the top of each section indicate the location of the maximum of A_1 for each sample [see Fig. 4(a)]. It can be seen that A_1 satisfies Eq. (3) for temperatures below the maximum, even for the most impure samples (such as CdAg-1) for which the impurity scattering takes control at a temperature somewhat lower than that where A_1 is a maximum.

Also plotted are curves that represent $10^3/r$ for the Cd group and for CdIn-C in the CdIn group. It is obvious that Eq. (3) holds for a large temperature range for samples which are not too impure and that the temperature dependence of A_1 is not the same as that for the electrical resistivity. In the high-temperature range ($T \gtrsim 200$ K), $A_1(T)$ seems to converge to $1/r(T)$.

A new characteristic temperature of the order of 43 K enters the picture. Several physical conditions could give rise to this behavior. The parameter could be a "virtual" temperature as a

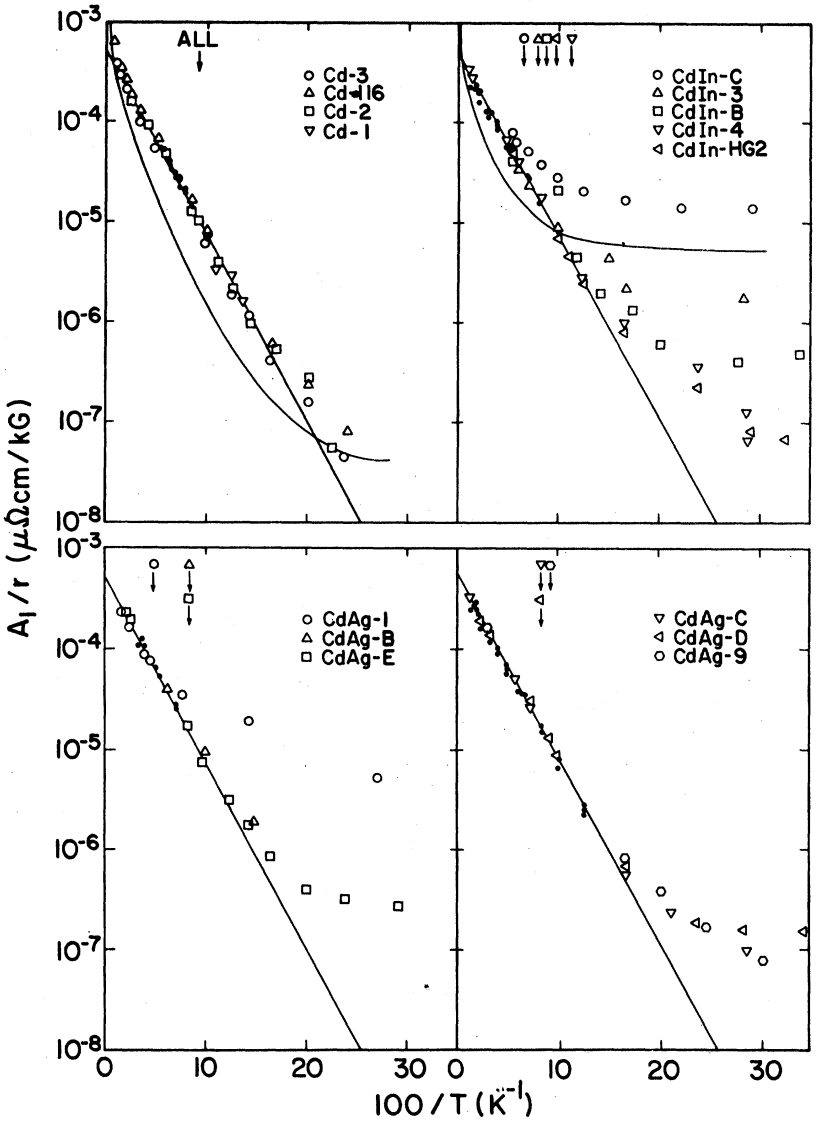

FIG. 5. Ratio A_1/r against $1/T$. The marks and arrows at the top of each section indicate the locations of the maximum of A_1 [Fig. 4(a)]. The two curves in the top sections represent $10^3/r$ for Cd-2 and CdIn-C, respectively. Single dots represent sample values where the large overlap of the marks would obscure their location.

result of interference between phonon modes, it could be the indication for a phonon scattering mode that dominates electron scattering in planes perpendicular to the principal axis, or it could indicate a continuous change in carrier density with decreasing temperatures. For a compensated metal the Hall resistivity contains terms of order greater than one in H, and the coefficient for the term in H does not necessarily reflect a property of the Fermi surface.[15] It seems in the present case that A_1 may reflect such a property in an indirect manner when, for example, a particular phonon mode could be found that interacts strongly with carriers in a certain portion of the Fermi surface.

One may question the significance of Eq. (3) in view of the fact that A_1 is a quantity that has been deduced from experimental data which are not, for every temperature, well established because in several cases A_1 had to be determined from measured $\rho_{21}(H)$ curves taken with only five field values. This objection carries weight; it means that a rather large uncertainty in the argument of the exponential in Eq. (3) can be expected when it is deduced from a single $\rho_{21}(H)$ curve. However, the fact that *all* the values of A_1 converge around Eq. (3) for temperatures above the impurity scattering range is rather convincing.

IV. RELATIONS THAT CONCERN ρ_{21m}

Considering that (for a given sample) ρ_{21m} occurs at a lower temperature for decreasing field values, the few fields H_m that were used in each case do indicate a possible exponential temperature dependence as illustrated for a few samples in a plot of $\ln H_m$ against $1/T_m$ (see Fig. 6, T_m is the tempera-

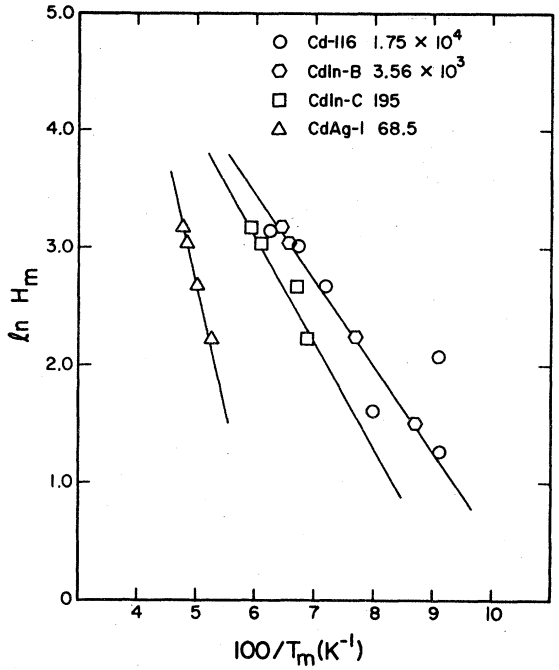

FIG. 6. Natural logarithm of the field H_m at which, for different temperatures, ρ_{21m} is observed against $1/T_m$.

ture for which ρ_{21m} in the field H_m is observed). A plot such as Fig. 6 does not make much sense. A plot of two particular variables (such as H_m and T_m) chosen at one characteristic value (such as ρ_{21m}) of a function of several dependent and possibly interdependent variables does not necessarily lead to relations that have physical meaning. On the other hand, one cannot exclude the possibility that such relations may provide pertinent information when they are combined with data from other experiments. For this reason and with the understanding that the approach is naive, the result of further analysis of H_m will be reported here.

It became clear that no generalized curve for ρ_{21m} against $(rH)_m \propto (\omega\tau)_m$ could be constructed. Quite a different result was obtained in a plot of $(H/r)_m$, which is equivalent to $(\omega/\tau)_m$, against T^{-1}. This is represented in Fig. 7, in which the $1/T$ axis is shifted to accomodate separately the different groups of samples. The straight lines represent the result for a multiple regression curve using all plotted data,

$$\ln (H/r)_m = (5.37 \pm 0.39) - (109.4 \pm 3.9)/T_m. \quad (4)$$

H_m is in kG, and Eq. (4) has a correlation factor equal to 0.97 with an average absolute deviation of 15% in $\ln (H/r)_m$ (which is indicated in Fig. 7 by the broken lines). The lower curve in the section for the pure cadmium samples represents $\ln (1/r_T)$ for Cd-2. Again caution is necessary because, as indicated before, no vigorous mathematical procedure was applied for finding the values for H_m. Therefore, maxima that could not be located within ± 0.2 K have been omitted. Furthermore, one may question the validity of any exponential behavoir that is restricted to a range of only a factor of 2 in one of the parameters (in this case temperature). On the other hand, the reliability of relation (4) is increased when one takes into account that it differs markedly from the nonlinear behavior of $1/r_T$, which in this temperature interval is determined with an error that is less than a few

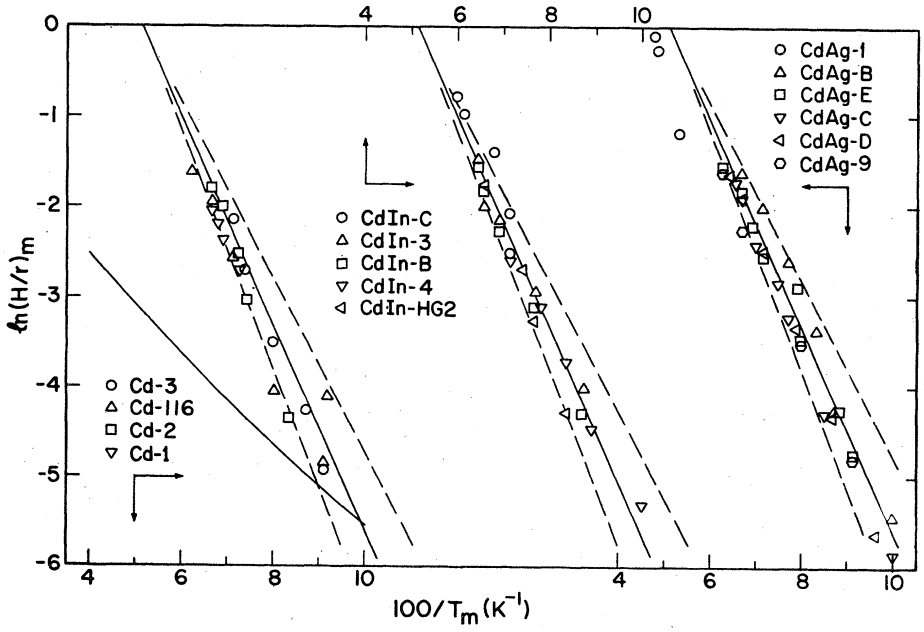

FIG. 7. Ratio $(H/r)_m$ against $1/T_m$. The broken lines represent 15% in the value of the ordinate. In the lower left corner the curve without marks represents $1/r_T$ for Cd-2.

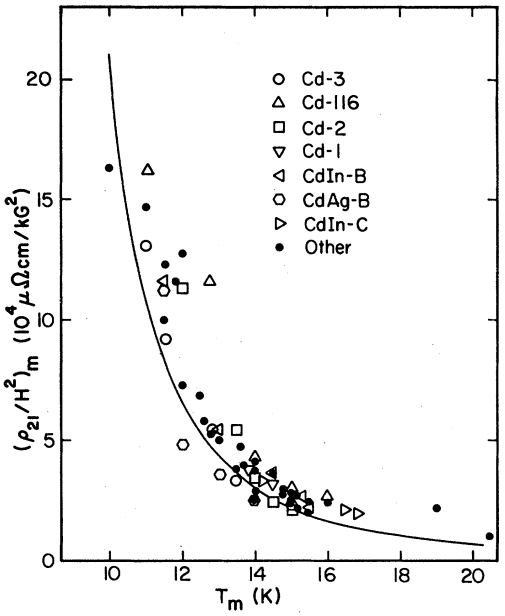

FIG. 8. Ratio between the measured values ρ_{21m} (at T_m) and the field squared against T_m. The curve represents the combination of the exponentials for $A_1/r(T)$ and $H_m/r_m(T)$.

percent. Also the difference in slope is a striking feature. The deviation from Eq. (1) that has been observed in ρ for a sample such as CdAg-1, with a relatively large impurity concentration, is also manifest in the relation for $(H/r)_m$. It is encouraging to find that at least the same temperature[16] is in the exponent of Eq. (4) as is in Eq. (1).

Accepting the expressions (3) and (4) at face value, one is tempted to combine them and compare the result with the experimental data in the temperature range $10 \leq T \leq 20$ K. Dividing Eq. (3) by Eq. (4) one obtains, considering that Eq. (3) holds for not too impure samples at temperatures including T_m and that $\rho_{21m} \approx (A_1 H)_m$ (see Fig. 4) at T_m,

$$A_1/H_m = (\rho_{21}/H^2)_m = 2.51 \times 10^{-6}\, e^{(66.5/T_m)}. \quad (5)$$

This reduced expression is plotted in Fig. 8 together with the measured quantities $(\rho_{21}/H^2)_m$ in a linear plot against temperature. The correlation is by no means good, but it is remarkable when one takes into account that Eq. (5) is the result of the manipulation of exponential functions with a reliability of the order of 80%. When all of the data $(\rho_{21}/H^2)_m$ (with the exception of a few that are rather far out) are analyzed, one finds $A/H_m = 5.5 \times 10^{-6}\, e^{59.5/T_m}$ with a correlation factor of 0.94.

V. SUMMARY AND CONCLUSIONS

The resistivity of pure Cd crystals and crystals with less than 50-ppm impurities seems to follow the Grüneisen-Bloch T^5 law, with a characteristic temperature $\Theta_R \approx 110$ K. This may indicate that the electrical conductivity in planes perpendicular to ⟨0001⟩ has a single carrier character without a large influence by umklapp scattering. A characteristic temperature $\Theta_R \neq \Theta_D$ is, in general, not uncommon, but it is of interest to note that a difference by a factor of 2 has been observed,[11] so far only for the case of Be.

The A_1/r relation is unexpected. Whatever its physical implication may be, this relation leads to an observation concerning the origin of ρ_{21m}. Except for the onset of impurity scattering, Fig. 5 indicates no deviations of the values A_1/r from Eq. (3) in the temperature range where ρ_{21m} occurs ($10 < T < 20$ K). This maximum has been studied rather thoroughly for the case of Cu. For Cu, a maximum occurs at temperatures around 40 K, and it is indicated by several authors[17,18] that the maximum is observed when H is parallel to a direction, such as ⟨211⟩, which is perpendicular to noninteracting open orbits. Two quite different interpretations of this orgin of ρ_{21m} in Cu have been suggested[19]: (a) an increase in $-\rho_{21}$ owing to the disappearance of umklapp processes in the neck when the temperature is lowered and (b) the changes with decreasing T, from certain orbits in low-field regime ($\omega\tau < 1$) to orbits with $\omega\tau > 1$, possibly coupled with a change in the balance of electron and hole states. In this present case $A_1 H$ is the dominant, and in particular for the pure samples the single, contribution to ρ_{21m}. It is difficult to understand that a low to high-field-regime exchange would pass unobserved in a plot such as Fig. 5, where A_1/r is a monotone function of T (for 5 K $\leq T <$ 300 K, which is independent of the estimate used for $\omega\tau$ at the temperature of the maximum.[20] This supports (at least for Cd) the fact that the origin of the maximum in ρ_{21} is to a large extent a process such as case (a), controlled by a change in the phonon scattering. This argument is enforced by the relations for $(H/r)_m$. It is difficult to entertain the possibility that the magnitude of $\omega\tau$ plays an unique role as the cause of ρ_{21m}. For example, at $T = 16.6$ K, ρ_{21m} is observed in a pure sample for which $(\omega\tau)_1 \propto (Hr)_m = 4 \times 10^3$ kG. For two much less pure samples CdIn-C and CdAg-1, values for $(Hr)_m$ of the order 1200 kG $\approx 0.3\,(\omega\tau)_1$ and 100 kG $\approx 0.025\,(\omega\tau)_1$, respectively, are obtained. For these samples differences of this magnitude in τ are acceptable. It is not acceptable to assume that in each case, within a temperature range of ± 5 K around 16 K, $\omega\tau$ should shift between low-field and high-field regimes. It would be also difficult to make acceptable the fact that the high-field regime could be reached in the case of the impure samples because of impurity generated orbits with a 40-times reduced cyclotron

hole mass.[21] Note[22] that for Cd, $m_e^*/m_h^* \lesssim 7$ for $H \parallel \langle 0001 \rangle$.

Finally, the following observation may be of interest for the physical interpretation of the presence of a strong phonon scattering that has been established in the present analysis. Numerical calculations have been made[23] on the phonon frequency distributions in Be, Mg, and Zn, and it is claimed that these give a fair representation of the experimental data that have been obtained by different methods. It was found that with increasing axial c/a ratios, the end point of the Debye range decreases rapidly in the following sequence (f in 10^{12} Hz): Be, $f \approx 11$; Mg, $f \approx 3.7$; and Zn, $f \approx 1.4$. In this same sequence Θ_D decreases from approximately 1150 to 400, to ~320 K and a rough extrapolation to 210 K for Cd yields an end-point frequency between 1.2×10^{12} and 0.6×10^{12} Hz. The temperature ($T \approx 11$ K, Fig. 4 (b)) that was found to appear for all samples implies according to a conventional estimate a dominant frequency $\approx 1.6\,kT/h \approx 1.2 \times 10^{12}$ Hz (which is of the right order of magnitude). Furthermore, the calculations[23] of the f distribution indicate that for Zn the end-point frequency 1.37×10^{12} Hz signals the onset of a large peak (up to 5×10^{12} Hz) with high intensity that is associated with an acoustic mode in the AH direction of the Brillouin zone. With this interesting deviation from the Debye spectrum as a starting point, it is indeed possible[24] to explain part of the relations that are reported in this paper.

ACKNOWLEDGMENTS

The author is very much indebted to Dr. D. A. Lilly, whose precise and extensive experiments are the basis for any measure of reliability that can be assigned to the derived relations. Thanks are due to Professor L. M. Falicov for his interest that was the encouragement needed for the publication of the result of the analysis, and also to M. C. Yoakam for the correction of several linguistic errors.

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*Work supported by the National Science Foundation Grants No. GP-12633A1 and No. DMR 73-02651.

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[19]This explanation and others are discussed with great care in Ref. 9.

[20]Assuming for pure Cd with $r_{4.2} \approx 30000$ an electron mean-free path of the order 1 mm [J. K. Galt, F. R. Merrit, and J. R. Klander, Phys. Rev. 139, A823 (1965)] and a Fermi velocity equal to 1.63×10^8 cm sec^{-1}, then $rH = 10^4$ kG corresponds to the free-electron value $\omega\tau = 3.6$. For actually longer mean-free paths $\omega\tau$ will increase proportionally, a similar change will occur for a cyclotron mass smaller than the free-electron mass.

[21]Whatever Eq. (5) means, it is clear from Fig. 7 that H_m/r_m decreases faster than $Hr^{-1}(T)$ in this temperature range. This difference suggests $H_m \propto \exp(-60/T_m)$. But see the remark made in Sec. IV on the physical significance of this kind of analysis.

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