temperature dependence of lattice thermal conductivity at liquid-He^3 temperatures.

Comparison of the temperature dependence of the two models is complicated by the fact that Gruner and Bross used three-phonon N processes instead of phonon-electron interactions to prevent low-frequency divergence. However, the two models also differ in other respects and lead to different temperature dependences. First, since the spacing of the dislocations forming the dipole must be less than the average distance between dislocations, deviations from the T^2 behavior should begin at a higher temperature. Second, for very long wavelengths in our model, the strained regions on either side of a single dislocation (compressed and dilated regions in the case of an edge dislocation) form a dipole which scatters as q_0^5, whereas the corresponding regions of a dislocation dipole form a quadrupole which scatters as q_0^7. Hence, the lattice thermal conductivity in the case of dipoles would change more rapidly away from a T^2 behavior than in our model, even if the same cutoff mechanism is invoked.

No measurements appear to have been made to date on the lattice thermal conductivity of deformed alloys below 1 °K. Such measurements would test the present theory and may provide information on dislocation densities.

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Knight Shift in Cadmium: Field and Temperature Dependence*

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Measurements have been performed of the Knight shift in cadmium as a function of field between 1 and 20 kG at 1.2 °K and as a function of temperature between 1.2 and 300 °K at 9.4 kG. The field-dependent studies show an oscillatory behavior of the Knight shift at the de Haas-van Alphen frequency appropriate to the first band in the higher-field ranges. In the low-field region the average value of the Knight shift is found to exhibit a strong anisotropic field dependence. The temperature-dependent measurements show that the anisotropy in the Knight shift reverses sign between 1.2 and 300 °K. Measurements of the amplitude of the de Haas–van Alphen signals were performed in the same field range and orientation as the oscillatory Knight-shift measurements. All of the measurements reported are accounted for by the detailed field and temperature dependence of the topology of the cadmium Fermi surface.

I. INTRODUCTION

There have been several measurements of the Knight shift σ in Cd over the past few years, but only recently have the measurements yielded consistent results and has a reasonable interpretation been given for them.[1] Cadmium is an interesting metal on which to compare measurements of σ with theory because the band structure[2] and particularly the Fermi surface[3] (FS) are extremely well known. In a series of recent papers, the results of field-dependent measurements of σ on single crystals of Sn,[4] Cd,[5] and Al[6] have been reported. It has been found that σ oscillates at the de Haas–van Alphen (dHvA) frequencies of electrons on the FS, and that through measurements of the amplitude of this oscillatory behavior one can determine wave-function amplitudes for selected groups of electrons on the FS. The wave-function amplitudes of conduction electrons can be determined through this technique at different points in the Brillouin zone once the FS and the amplitude of the dHvA signals are known. The purpose of this paper is to report on field- and temperature-dependent measurements of σ in Cd from which information about the conduction-electron wave functions can be obtained.

The possibility of dHvA-type oscillations in σ was originally suggested by Das and Sondheimer.[7] They conjectured that since

$$\sigma = \tfrac{8}{3}\pi\chi_p \langle |\psi(0)|^2 \rangle_{E_F}\Omega = \tfrac{8}{3}\pi\chi_p p_F\Omega \qquad (1)$$

[where χ_p is the conduction-electron spin susceptibility, Ω the atomic volume in which the electronic wave functions are normalized, and $\langle |\psi(0)|^2 \rangle_{E_F} = p_F$ the average conduction-electron density at the nucleus for electrons on the FS], σ should oscillate at the dHvA frequency because χ_p is oscillatory. Later, a calculation was performed by Stephen[8] using the scalar effective-mass approximation in which both the effects of the Pauli-spin paramagnetism and the diamagnetism of the conduction-electron system were included. Following Sn I, Glasser[9] gave a theory for the oscillations in σ in which the possibility of oscillations in p_F was allowed and this led to somewhat larger amplitudes of the oscillations than that found by Stephen. Weinert and Schumacher[10] pointed out that one cannot consider the paramagnetic and diamagnetic components of σ independently, but that they must be added with the appropriate phases of the oscillations to give the correct result. They further showed that the free-electron value of $p_F = 1/\Omega$ grossly underestimates the value of p_F and that the use of experimentally determined values of p_F leads to better agreement with Stephen's theory. For a discussion of the details of each of the theoretical treatments of $\tilde{\sigma}$, the oscillatory component of σ, the reader is referred to Sn II. The method of analysis used in the present work is given in detail in Ref. 6.

Recently Kasowski and Falicov[1] have explained the peculiar behavior of the temperature dependence of the anisotropy of σ in Cd by including the effects of the modulation of the pseudopotential coefficients caused by the lattice vibrations into the band calculation of Stark and Falicov.[2] Values for the isotropic and anisotropic parts of σ were calculated for temperatures between 0 °K and the melting point of Cd. The theoretical calculations give excellent agreement with the present temperature-dependent measurements.

During the course of investigating the oscillatory behavior of σ, it was noticed that the average value of σ was slightly field dependent. Consequently, a field-dependent study of σ between 1 and 20 kG with the field applied parallel and perpendicular to the hexagonal axis was performed. These results are presented and discussed here.

In the sections which follow, a description of the experimental procedure for the σ and dHvA measurements is given, the experimental results are presented, and a discussion of the results is given.

II. EXPERIMENTAL DETAILS

All of the samples used in this experiment were cut by spark erosion from a zone-refined bar of Cd ($R_{300\,°K}/R_{4.2\,°K} \approx 21\,000$) obtained from Cominco Products Inc. (quoted purity 99.9999%). The sample used in the field-dependent measurements was a series of thin wafers interleaved with Mylar and prepared in a manner similar to that described for Sn in Sn II. Since it is known from the dHvA work of Tsui and Stark[11] that a maximum in amplitude of the dHvA signal and minimum in area occurs for the first-zone caps at an angle of 30° from the [0001] axis in the $(10\bar{1}0)$ plane, each wafer was cut such that this direction was contained in the plane of the wafers. The average thickness of each wafer was 0.3 mm and the over-all size of the sandwiched sample was 14×10×10 mm.

Measurements of the anisotropic Knight shift were made on a cylindrical monocrystal cut from the same starting material as the wafers. The axis of the cylinder was along the $[10\bar{1}0]$ direction so that the field $\vec{H}$ could be rotated perpendicular to this axis from the [0001] to the $[11\bar{2}0]$ direction. About 20 evenly spaced radial slots were cut in the sample to increase the exposed surface area. The final outside dimensions of the cylinder were a 5-mm radius and a 15-mm length.

The method of recording the NMR lines at each field value was the same as that described in Sn II; however, for Cd the method of analyzing the data was somewhat different. The true center of each of the recorded NMR lines was obtained by correcting the apparent center (midway between the posi-

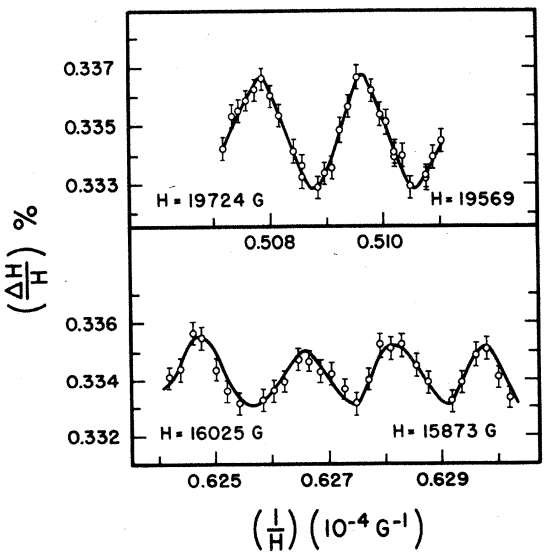

FIG. 1. Experimental values of the oscillation in σ as a function of inverse field. The points shown in the figure have been corrected for line-shape distortion and the error bars represent the total uncertainty in determining the true center of the NMR line at each value of the field.

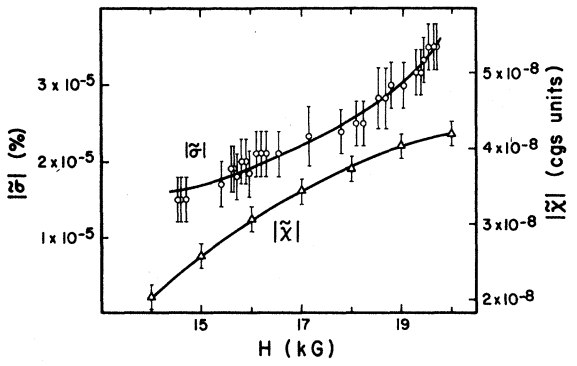

FIG. 2. Amplitude of $\tilde{\sigma}$ and $\tilde{\chi}$ as a function of applied magnetic field. Each of the points on these curves represents an average over approximately a 100-G range. The $|\tilde{\chi}|$ results have been corrected for the field gradient in the calibration measurements.

tions of maxima and minima of the derivative curve) for eddy-current distortion,[12] and this corrected line center was used to obtain values of the Knight shift at each field and angle. The Cd^{113} NMR signal from a saturated solution of $CdCl_2$ doped with a 0.1-mole% addition of $MnCl_2$ was used as a reference to calculate σ. The amount of line distortion was slightly dependent on the crystallographic direction in which the field was applied with the line slightly more distorted for the field along [0001] than along $[11\bar{2}0]$. The distortion correction was applied to calculate anisotropies as well as field-dependent effects.

As in the work on Sn in Sn II, measurements were made of the dHvA susceptibility on the same samples as the Knight-shift measurements were performed. The field-modulation technique was used for extended-field-range measurements of the amplitude of the oscillations. In these measurements a modulation frequency of 135 Hz and modulation amplitude of 2 G peak to peak was used. In order to obtain a calibration of the amplitude of the susceptibility oscillations, the field dependence of the susceptibility of one of the wafers was measured in a force magnetometer.[13]

III. EXPERIMENTAL RESULTS

In the range of fields between 12 and 20 kG with $\vec{H}$ applied 30° from the [0001] axis in the $(10\bar{1}0)$ plane, the Knight shift in Cd is found to oscillate at the dHvA frequency appropriate to the first-zone caps. This result is displayed in Fig. 1 for two ranges of magnetic field covering about 150 G in each case. The upper part of this curve shows a different scale from that in Ref. 5. This is due to a change in magnet calibration which occurred during the course of this investigation and has subsequently been corrected. Each of the NMR lines used to calculate σ for the points on the curve was corrected for line-shape distortion.[12] The distortion correction only leads to a reduction in scatter of the points and the oscillating Knight shift is easily discernible when the uncorrected line centers are used to calculate σ vs H.

The average peak-to-peak amplitude of three to four of the oscillations was measured at various values of field and plotted versus H. This result is shown in Fig. 2 along with the average amplitude of the oscillations in the susceptibility χ vs H for the same field direction. The force magnetometer used to calibrate the amplitude of the dHvA oscillations in χ had a field gradient of 600 G/cm at 20 kG perpendicular to the applied field direction. The sample used for the calibration was 0.25 mm thick in the direction of the gradient, which made the gradient across the sample 15 G. When this gradient is compared to the 75-G period of the first-zone caps at 20 kG, the gradient reduces the amplitude by about 20% due to the change in phase of the dHvA frequencies over the sample.[14]

The Knight shift at a constant field of 9.4 kG was measured for several temperatures with $\vec{H}$ both parallel and perpendicular to [0001] in the solid cylindrical sample and these results are shown in Fig. 3. The interesting part of these curves is that the anisotropy reverses sign between 35 and 40 °K. These results will only be discussed briefly since they have been analyzed in detail by Kasowski and Falicov.[1]

Measurements of σ with the field parallel to the c axis, $\sigma_{\parallel}$, and perpendicular to the c axis, $\sigma_{\perp}$, were made from 1 to 18 kG. This result is shown in Fig. 4 and shows that $\sigma_{\parallel}$ decreases rapidly as the field is decreased below 5 kG and that $\sigma_{\perp}$ remains rela-

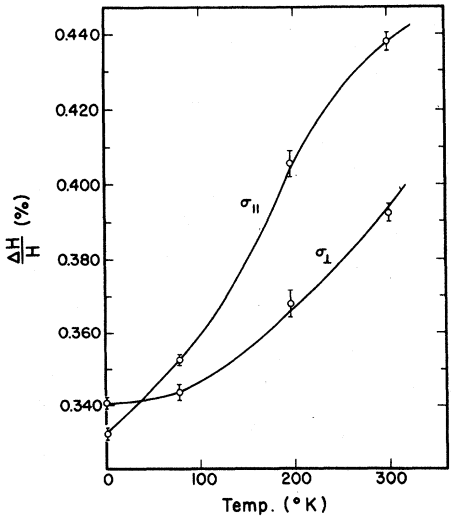

FIG. 3. Temperature dependence of the Knight shift in a single crystal of Cd for the magnetic field directed both parallel and perpendicular to the hexagonal axis.

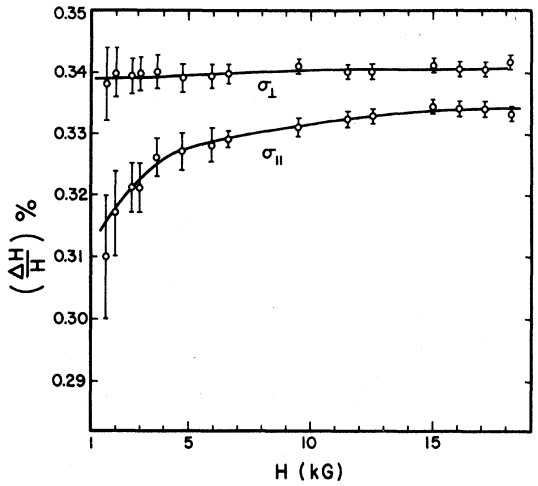

FIG. 4. Field dependence of the average value of σ for $\vec{H}$ directed parallel to the [0001] axis ($\sigma_{||}$) and parallel to the [11$\bar{2}$0] axis ($\sigma_\perp$). Below 10 kG, the error bars represent the maximum scatter for between six to eight measurements at the same field for two different samples. Above 10 kG, the error bars represent the amplitude of $\tilde{\sigma}$.

tively independent of field for the entire field range. All of these measurements were performed on the solid cylindrical sample. For the field directed along the c axis, the average value of σ continues to increase linearly to 20 kG.

IV. DISCUSSION OF RESULTS

A. Amplitude of Knight-Shift Oscillations

Before proceeding to a discussion and comparison of the data with various theories of $\tilde{\sigma}$, there is one possible mechanism which might give rise to dHvA oscillations in the position of the NMR line and it needs to be discussed. Since a nucleus in the metal is acted upon by the B field existing in the metal, the oscillatory magnetization in the sample would change the NMR line position at the dHvA frequency. In fact, all of the theoretical treatments of σ should use $B = H + (4\pi - D)M$, where M is the magnetization and D the demagnetizing factor of the sample in the calculations. Thus, the expression for σ should be written

$$\sigma = \frac{\Delta B}{B_0} = \frac{\Delta[H + (4\pi - D)M]}{H_0 + 4\pi M_0} = \frac{\Delta H}{H_0} + \frac{(4\pi - D)M}{H_0}, \quad (2)$$

where the magnetization of the reference compound M_0 is assumed to be negligibly small and can be easily checked by using different reference compounds to measure σ.

The amplitude of the oscillatory magnetization $\tilde{M}$ in the wafers was measured as a function of applied $\vec{H}$ and the results given in Fig. 2. As can be seen from Fig. 2, $(4\pi - D)\tilde{M}$ is less than $\frac{1}{10}$ the amplitude of $\tilde{\sigma}$, in addition to the fact that the field dependence of the two amplitudes is different. It is concluded, therefore, that although $\tilde{M}$ may contribute to the amplitude of $\tilde{\sigma}$, more than 90% of this amplitude arises from other factors.

The effect of sample magnetization is usually not considered in Knight-shift measurements. This practice may, however, lead to considerable error in two cases: (i) when σ is very small as in Be, and (ii) when the anisotropic part of σ is being calculated. The latter error may be large because this calculation involves taking a difference in measurements of σ along two directions and the susceptibility may be anisotropic as in the case of Cd.[15]

B. Oscillatory Knight Shift

The first analysis that was performed on the oscillatory-Knight-shift data was a nonlinear least-squares fit to the expressions given by Stephen.[8] His expressions for both the paramagnetic and diamagnetic components were added with a variable phase difference between them and used in the fit. The fit was performed in several field ranges with three adjustable parameters: the amplitudes of the two components and the phase between them. The data could be fitted in this manner with a correlation of 0.98. This calculation shows that the diamagnetic contribution of $\tilde{\sigma}$ is less than 10% of the paramagnetic contribution in the field range 16–20 kG and, hence, will not be discussed further.

The difficulty with this type of analysis is that the amplitude of the paramagnetic component involves the product of the oscillatory density of states and P_F. A different procedure for analysis of the data has been given in detail by Khan *et al.*[6]. Here the amplitude of the oscillatory density of states is computed from measurements of the dHvA amplitude and the many-body effects scaled out by taking the ratio of $\tilde{\sigma}$ to σ. Thus, the ratio

$$\frac{\tilde{\sigma}}{\sigma} = \frac{\tilde{\chi}_\rho}{\chi} \frac{\langle |\psi(0)|^2_{E_F} \rangle_{\text{oav}}}{\langle |\psi(0)|^2_{E_F} \rangle_{\text{av}}}$$

is computed.[5] In this expression $\langle |\psi(0)|^2_{E_F} \rangle_{\text{oav}}$ is the average value of P_F over the particular orbit on the FS contributing to the oscillations. As was pointed out in Ref. 6, the cancellation of the many-body effects may not be complete since it is tacitly assumed that the mass enhancement in the diamagnetic susceptibility is the same as it is in the spin susceptibility.[16] Using this analysis,

$$\langle |\psi(0)|^2_{E_F} \rangle_{\text{oav}} / \langle |\psi(0)|^2_{E_F} \rangle_{\text{av}} = 1.67$$

is obtained for the orbit observed on the first-band caps in Cd.

C. Temperature Dependence of $\sigma_{||}$ and $\sigma_\perp$ in Cd

The experimental values of the temperature dependence of $\sigma_{||}$ and $\sigma_\perp$ are presented in Fig. 3. Here $\sigma_{||}$ is the value of σ with $\vec{H}$ directed along the [0001] direction in the crystal and $\sigma_\perp$ is for $\vec{H}$ along

the [11$\bar{2}$0] direction. The present results are in particularly good agreement with a previous measurement of $\sigma_{\parallel}$ and $\sigma_{\perp}$ in Cd by Borsa and Barnes.[17] As the temperature is decreased from 300 to 77 °K, $\sigma_{\parallel}$ decreases faster than $\sigma_{\perp}$, and below 77 °K, $\sigma_{\perp}$ does not change significantly while $\sigma_{\parallel}$ shows a continuous decrease at an almost constant rate. The values of σ given in Fig. 3 have not been corrected for the susceptibility contribution to σ.

A measure of the temperature dependence of the anisotropy of the charge distribution at the Fermi surface in Cd is obtained through the difference in $\sigma_{\parallel}$ and $\sigma_{\perp}$. Since $\sigma_{\parallel} = \sigma_{iso} + \sigma_{an}$ and $\sigma_{\perp} = \sigma_{iso} - \frac{1}{2}\sigma_{an}$, where σ_{iso} is the isotropic part of σ and σ_{an} the anisotropic part of σ, $\frac{3}{2}\sigma_{an}$ is obtained by $\sigma_{\parallel} - \sigma_{\perp}$. The result for Cd is

$$\sigma_{\parallel} - \sigma_{\perp}/\sigma_{iso}\, T = 300\,^{\circ}\mathrm{K} = 11.9\%,$$

$$\sigma_{\parallel} - \sigma_{\perp}/\sigma_{iso}\, T = 4.2\,^{\circ}\mathrm{K} = -3.0\%,$$

showing that between 300 and 4.2 °K the charge distribution is considerably changed with an isotropic distribution occurring between 35 and 40 °K.

The nonlocal pseudopotential calculation of the energy bands in Cd by Stark and Falicov[2] shows that there is a rather strong influence on the electrons at the Fermi surface by the *d* bands. This fact causes departures of the Fermi surface from the free-electron surface in a nonlocal manner. Thus, the third-band lens-shaped portion of the surface centered at the point Γ in the Brillouin zone remains practically unchanged from the free-electron surface while the second-band surface centered along the *KH* direction is about one-half the free-electron size and distorted from the free-electron shape. These various parts of the Fermi surface will contribute to σ in different ways and the detailed temperature dependence of the pseudopotential coefficients must be calculated to obtain agreement with experiment.

Calculations of σ including these effects have been performed by Kasowski and Falicov[1] and in more detail by Kasowski.[1] Their results show very good agreement with the present measurements and point out the importance of the symmetry character of the wave functions of electrons on different parts of the FS to their contribution to σ.

D. Field Dependence of Average Value of σ

If the total contribution to σ were that given by Eq. (1), σ would be field independent for all values of $\vec{H}$ except for the oscillatory behavior. The data presented in Fig. 4, however, show that this is not the case for $\vec{H}$ directed parallel to the [0001] axis. A field-dependent behavior of σ of this magnitude can be caused by several effects. A large anisotropic diamagnetic contribution could give rise to the observed field dependence. To our knowledge, there is currently no theory which gives expressions for σ in the presence of strong diamagnetic effects and the available measurements of the diamagnetic susceptibility cannot account for this change. In what follows, a description of the difference in orbit geometry on the second-zone hole portion of the FS for the two different field orientations is given. Differences in scattering mechanisms for orbits in the two field directions are pointed out and shown to lead qualitatively to a plausible explanation of the data.

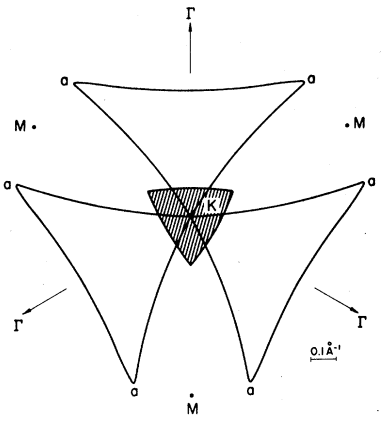

FIG. 5. Outline of the Fermi surface in the second zone of Cd. The large trifoliate outline lies in the ΓMK plane and the cross-hatched triangular outline lies in the *ALH* plane.

A complete description of the second-zone sheet of the FS is given in Ref. 3. Figure 5 is a projection of the extremities of the surface onto the ΓMK plane. The large trifoliate shape outline centered at the *K* point lies in the ΓMK plane and the cross-hatched triangular section lies in the *ALH* plane with its center at *H*. Since magnetic breakdown effects for the field directed along [0001] do not become appreciable until well above 20 kG,[11] the behavior between 1 and 10 kG reported here cannot be accounted for by breakdown.

A mechanism which could contribute to the observed field dependence is intrasheet scattering between adjacent arms of the trifoliate structure at the point labeled *a* in Fig. 5. This scattering causes the electron orbits to remain in the same Brillouin zone in the repeated zone scheme. The effect of this scattering process was first observed by Grenier *et al.*,[18] who measured a reversal of the Hall coefficient as a function of field when $\vec{H}$ was applied parallel to [0001]. The scattering machanism responsible for the effect was first proposed by Young, Ruvals, and Falicov[19] and schematically is as follows. Owing to the flatness of the second energy band along the ΓM line near the Fermi energy, the probabilility of tunneling across this line is much greater than at other

points on an electron's trajectory on the FS. Thus, a high probability of tunneling from one trifoliate sheet to the next occurs at this point. As the magnetic field increases, the tunneling probability is decreased and the orbits for the field directed along [0001] return to their trifoliate shape.

Thus, one sees that as the field increases at constant temperature, the orbits on the second-zone hole surface change from electronlike to holelike. This is what causes the reversal in the sign of the Hall coefficient as a function of field for this orientation between 0 and 10 kG. If the symmetry of the wave functions of these tunneling electrons changes from p to s due to this process, then σ will change as a function of $\vec{H}$. For the field directed along the $[11\bar{2}0]$ direction, no orbits intersect the tunneling point and the symmetry of the electrons on these orbits remains unchanged for all fields investigated here. At fields greater than 20 kG, magnetic breakdown along the AH line and at the K point should give rise to a change in σ for the same reasons.

V. CONCLUSIONS

The basic conclusion to be drawn from the series of Knight-shift measurements reported here is that in order to correctly predict the behavior of σ under varying experimental conditions, great attention must be paid to both the details of the FS on which the electrons move and the various scattering mechanisms in which they are involved. Thus, the oscillatory behavior is accounted for by detailed considerations of the magnitude of the s-type character of the extremal orbits giving rise to the oscillations. The temperature dependence is explained by the changes which occur in the band structure due to modulation of the lattice potential by the temperature. And, finally, the particular scattering or tunneling mechanisms active on the electronic orbits can cause the value of σ to change considerably.

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