framework of the present model. However, the suggestion that has been made does seem to offer a plausible qualitative explanation for the systematic character of the discrepancies identified in both Tables I and II.

In summary, it has been shown that the inclusion of lattice distortion much improves agreement between experiment and theory for the residual resistivity of zinc alloys. Thus, although systematic discrepancies have been identified and discussed, we conclude that significant lattice-distortion screening occurs in zinc-based alloys.

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†Present address: Physics Department and Institute of Materials Science, University of Connecticut, Storrs, Conn. 06268. Partially supported by AFOSR 68-1517.

‡Present address: Houghton College, Houghton, N.Y.

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Effect of Thermal Umklapp on the Low-Temperature Electrical Resistivity

A. C. Ehrlich

Naval Research Laboratory, Washington, D.C. 20390

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The Klemens-Jackson theory of the low-temperature resistivity of those monovalent metals whose Fermi surface touches or nearly touches the Brillouin-zone boundary has been extended to include the presence of impurity scattering, which is described by an isotropic relaxation time. The electrical resistivity thus obtained is in the form of a slowly converging infinite series. The deviations from Matthiessen's rule which occur as a consequence of the simultaneous presence of thermally induced umklapp processes and impurity scattering are calculated. They prove to be large and have a temperature dependence that, over limited temperature intervals, could be spuriously interpreted as showing evidence of electron-electron scattering.

In recent years, there has been considerable interest in the temperature dependence of the electrical resistivity at low temperatures. This interest has arisen because the low-temperature electrical resistivity can, in principle, indicate the presence or absence of certain electron-electron interactions. Baber scattering,[1] spin density fluctuation scattering,[2] and magnon scattering[3] can be expected to occur in transition, nearly ferromagnetic, and magnetic metals, respectively, and will give rise to a T^2 term in the electrical resistivity, where T is the absolute temperature. Very recently it has been shown that the electrical resistivity in some nontransition metals also can be separated into a residual, a T^2, and a T^5 term.[4]

In order to attribute a distinct scattering mechanism to each of the various terms into which the experimental resistivity data are separated, it is necessary to assume the validity of Matthiessen's rule. While such an assumption is well known to be imprecise, the absence of any realistic alternative procedure makes this assumption necessary. In the present paper, we have extended the theory of Klemens and Jackson[5] (KJ) of the low-temperature resistivity in simple metals in which the authors assumed that only phonon scattering is present. We have treated the problem in an entirely analogous way, except that we have allowed an arbitrary amount of impurity scattering to be acting simultaneously with the phonon scattering. The resulting deviations from Matthiessen's rule are shown to be large (in the sense that the temperature-dependent part of the resistivity deviates strongly from what would be expected) and in a manner which could easily be confused with an electron-electron interaction term.

The theory in KJ applies to a simple metal whose (spherical) Fermi surface touches or nearly touches the Brillouin-zone boundary. In this situation, low-temperature thermally induced umklapp processes can occur around this area of contact (for example, around the necks on the Fermi surface of the noble metals). Use is made of the fact that the electron-phonon interaction at low temperatures moves an electron over the Fermi surface (assumed spherical) in random steps sufficiently small that the process can be regarded as taking place by diffusion. The electrical field E_z (taken in the z direction) is treated as a spatially distributed "source" of electrons. Since $E = E_z$, the spatial distribution is equal to $\alpha\cos\theta$, where $\alpha = (eE_z/\hbar)(\partial f_0/\partial k)$ and $\theta(\varphi)$ is the polar (azimuthal) angle. Boltzmann's equation then becomes a two-dimensional diffusion equation [see KJ, Eq. (3)].

Klemens and Jackson[5] solved such an equation for a simple cubic lattice with the boundary condition that the deviation from equilibrium of the Fermi function g is zero at $\theta = \epsilon$, $\theta = \pi - \epsilon$. This boundary condition corresponds to contact of the Fermi sphere with the Brillouin-zone boundary at these two values of θ. The other four areas of contact at $\theta = \frac{1}{2}\pi$, $\varphi = 0$, $\frac{1}{2}\pi$, π, $\frac{3}{2}\pi$ are neglected, since g vanishes there in any case.

We account for the impurity scattering by supposing that it can be described by an isotropic relaxation time τ_i in the usual way. This means that in the absence of thermal umklapp scattering, Matthiessen's rule would be obeyed. Noting that g is not a function of φ by symmetry, in the simple cubic case, and letting $\mu = \cos\theta$, our Boltzmann equation takes the form

$$(1-\mu^2)\frac{\partial^2 g}{\partial\mu^2} - 2\mu\frac{\partial g}{\partial\mu} - \frac{k_F^2}{D}\frac{g}{\tau_i} = -\alpha\mu\frac{k^2}{D} \quad , \qquad (1)$$

where k_F is the radius of the Fermi sphere and D the diffusion coefficient. We shall designate $k_F^2/2D$ as τ_p, the phonon relaxation time, since it can easily be shown that in the absence of zone-boundary–Fermi-surface contact, we obtain $k_F^2/2D = \tau_p$. A solution to the inhomogeneous equation (1), g^i, is

$$g^i = \alpha[\tau_i\tau_p/(\tau_i+\tau_p)]\,\mu = \alpha\tau\mu \quad , \qquad (2)$$

where $1/\tau = 1/\tau_i + 1/\tau_p$.

The remaining homogeneous equation can be written

$$(1-\mu^2)\frac{\partial^2 g}{\partial\mu^2} - 2\mu\frac{\partial g}{\partial\mu} - Pg = 0 \quad , \qquad (3)$$

where $P = 2\tau_p/\tau_i$.

Equation (3) closely resembles Legendre's equation, differing only in the sign of the third term. The general solution of Legendre's equation is a sum of two infinite series, one odd and the other even, in μ.[6] Calling these odd and even series s and t, respectively, the solution of Legendre's equation y can, in fact, be written

$$y = As(\mu, P) + Bt(\mu, P) \quad , \qquad (4)$$

where A and B are undetermined constants. It follows that the general solution to Eq. (3) is

$$g^h = As(\mu, -P) + Bt(\mu, -P) \quad . \qquad (5)$$

Since the general solution of Eq. (1) is the particular solution [Eq. (2)] plus the general solution of the homogeneous equation [Eq. (5)], we have

$$g = \alpha\tau\mu + As(\mu, -P) + Bt(\mu, -P) \quad . \qquad (6)$$

A and B are determined by applying the same boundary conditions as Klemens and Jackson,[5] i.e.,

$$g = 0, \qquad \theta = \epsilon, \qquad \theta = \pi - \epsilon. \qquad (7)$$

These give

$$A = -\alpha\tau\eta/s(\eta, -P), \quad B = 0, \qquad (8)$$

where $\eta = \cos\epsilon$. Now,

$$s(\mu, -P) = \sum_{n=0}^{\infty} R_{2n+1}\mu^{2n+1} \quad , \qquad (9)$$

with

$$R_{2n+1}=\prod_{m=1}^{n}\left[P+2m(2m-1)\right]\frac{1}{(2n+1)!}\,,\quad n>0 \tag{10}$$

$$R_1=1,\qquad n=0.$$

Combining Eqs. (6), (8), and (9), we obtain

$$g=\alpha\tau\mu-\alpha\tau\eta\sum_{n=0}^{\infty}R_{2n+1}\mu^{2n+1}\Big/\sum_{n=0}^{\infty}R_{2n+1}\eta^{2n+1}\quad . \tag{11}$$

Now,

$$\begin{aligned}j&=\frac{2e}{(2\pi)^3}\frac{\hbar}{m}\int_0^{k_F}\int_0^{2\pi}\int_0^{\pi}k^3 g\cos\theta\,\sin\theta\,d\theta\,d\varphi\,dk\\&=\tfrac{4}{3}\pi\alpha'\tau\eta^3\left[1-\left(\sum_{n=0}^{\infty}\frac{3}{2n+3}R_{2n+1}\eta^{2n}\Big/\sum_{n=0}^{\infty}R_{2n+1}\eta^{2n}\right)\right],\end{aligned} \tag{12}$$

where $\alpha'=2(k_F/2\pi)^3(e^2/m)E_z$, and e and m are the electronic charge and mass, respectively. For an equivalent spherical Fermi surface not in contact with a Brillouin-zone boundary, the current is

$$j=\tfrac{4}{3}\pi\alpha'\tau\quad .$$

The physical significance of the various terms in Eq. (12) is, therefore, that η^3 represents the reduction in current that arises when a section of the Fermi surface is in contact with the Brillouin-zone boundary. Such a section cannot contribute to the current, as is well known. The quotient of the two infinite series gives the reduction in current caused by the umklapp processes and depends only on the Fermi-sphere–Brillouin-zone contact area (through η) and P, twice the ratio of the residual resistivity to the thermal resistivity *in the absence of umklapp processes*. These series converge rather slowly if η is near unity and thus can be accurately evaluated only by electronic computer.

In order to compare the temperature-dependent resistivity predicted by the present calculation with that obtained when Matthiessen's rule holds, it is convenient to designate

$$G(\eta,P)=\sum_{n=0}^{\infty}\frac{3}{2n+3}R_{2n+1}\eta^{2n}\Big/\sum_{n=0}^{\infty}R_{2n+1}\eta^{2n}\quad .$$

Using $\sigma=j/E_z$ and Eq. (12), we may write

$$\sigma=\sigma'(1-G),\quad \rho=\rho'[1/(1-G)]\quad , \tag{13}$$

where $\sigma(\rho)$ is the conductivity (resistivity) calculated here and $\sigma'(\rho')$ the conductivity (resistivity) in the absence of umklapp processes. By hypothesis, ρ' can be written as the sum of the residual and thermal resistivities

$$\rho'=\rho_0'+\rho'(T) \tag{14}$$

and $\rho_0'=\rho_0,\quad P=2\rho_0'/\rho'(T)\quad .$ (15)

So, combining Eqs. (13)–(15)

$$\frac{\rho-\rho_0}{\rho'(T)}=\frac{P}{2}\left(\frac{1}{1-G}-1\right)+\frac{1}{1-G}$$

$$=F(P,\eta) \tag{16}$$

or $\rho-\rho_0=\rho(T)=\rho'(T)F(P,\eta)\;.$ (17)

Equation (17) states that the temperature variation of resistivity when umklapp scattering processes are absent, $\rho'(T)$, must be modified by the multiplicative function $F(P,\eta)$ when these processes are present. Since F is a function of temperature, through its dependence on P, $\rho(T)$ will not have the same functional dependence on temperature as $\rho'(T)$.

The procedure to obtain corresponding solutions for the fcc and bcc crystal structures is similar to that used in KJ. As was done there, we can obtain approximate solutions using a superposition of the previously obtained solutions centered about axes through the various regions of Fermi-surface–zone-boundary contact (i.e., necks). In fact, certain approximations, valid in the absence of impurity scattering, involving variation of the solutions centered about one neck over the perimeter of another neck, are even better when impurity scattering is present. To illustrate the procedure for the fcc structure (applicable to the noble metals), we take $\vec{E}$ in the [100] direction and try a solution of the form

$$g=\alpha\tau\mu+\alpha\tau\sum_{i=1}^{4}Cg_{2i}(\mu_i,P)\;, \tag{18}$$

where $g_2=\sum_{n=0}^{\infty}R_{2n+1}\mu^{2n+1}\quad .$

The condition that g vanishes on the base of one of the necks is

$$O=1/\sqrt{3}+Cg_2(\eta,P)+Cg_2(\tfrac{1}{3},P)$$

or $$C=-\frac{1}{\sqrt{3}}\times\frac{1}{g_2(\eta,P)+g_2(\frac{1}{3},P)}\quad . \tag{19}$$

[See KJ, Eqs. (21) and (22) for comparison and details.]

Using Eqs. (18) and (19) gives

$$g=\alpha\tau\mu-\alpha\tau(4/\sqrt{3})\sum_{n=0}^{\infty}R_{2n+1}\mu^{2n+1}\times\left(\sum_{n=0}^{\infty}R_{2n+1}\eta^{2n+1}+\sum_{n=0}^{\infty}R_{2n+1}(\tfrac{1}{3})^{2n+1}\right)^{-1}\quad . \tag{20}$$

The procedure in going from Eq. (20) to an expression for the temperature-dependent resistivity is entirely equivalent to that employed in the development from Eq. (11) to Eq. (17). The result appears identical to Eq. (17), but $F(P,\eta)$ is a different function. A procedure analogous to this also

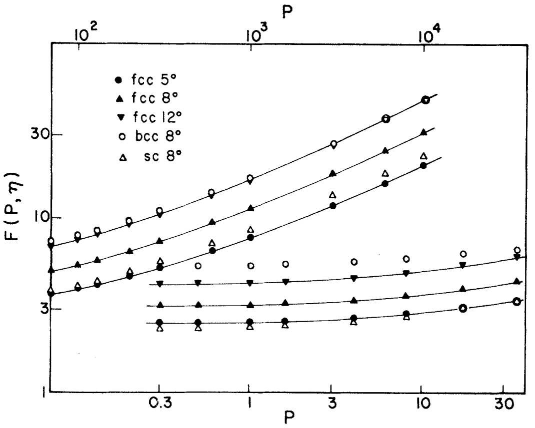

FIG. 1. A logarithmic plot of $F(P, \eta)$ versus P for fcc crystals with $\epsilon = 5°$, 8°, and 12° and for bcc and simple cubic crystals with $\epsilon = 8°$. The upper and lower abscissas correspond to the upper and lower curves, respectively. For clarity, curves have been drawn through the fcc results.

has been carried through for the bcc structure. In both of these cases, just as for the simple cubic crystal symmetry, numerical results can be obtained only through the use of a high speed computer to evaluate the rather slowly converging infinite series that comprise the function $F(P, \eta)$. These computer calculations have been performed for the three cubic crystal structures for a large number of values of η.

In Fig. 1 is presented a logarithmic plot of F versus P for the fcc crystal structure for three values of η corresponding to $\epsilon = 5°$, 8°, and 12°. For comparison we also show the results for bcc and simple cubic lattices with $\epsilon = 8°$. There are differences in detail, but all the curves are approximately parallel to each other.

The magnitude of F in Fig. 1 indicates that $\rho(T)/\rho'(T) > 1$, having the value given by Klemens and Jackson[5] for $P \lesssim 1$. The slope of $\ln F$ as a function of $\ln P$ is not constant, but varies smoothly between approximately 0 for $P \lesssim 1$ to 0.5 at $P = 10^4$. Since $P \propto (1/T)^5$ [if $\rho'(T)$ is assumed to vary as T^5] it follows from Eq. (17) that $\rho(T) \propto T^5$ for $P \lesssim 1$, but will have no simple variation with temperature for $P > 1$.

The shape of the curves in Fig. 1 suggests that over limited temperature intervals the temperature dependence of ρ can have various functional dependencies of the form $\rho \propto T^n$. In fact, for *any* η and P considered, ρ can be shown to fit an expression of the form

$$\rho = \rho_0 + AT^2 + BT^5 \quad . \qquad (21)$$

This has been done in Fig. 2, where $(\rho - \rho_0)/T^2$ has been plotted as a function of T^3 for the $\epsilon = 5°$, 8°, and 12° fcc calculations. The units of temperature and resistivity are arbitrary. What is significant is that P covers the range 10^4–4, a factor of 2.5×10^3, which corresponds to a variation in temperature of a factor of $(2.5 \times 10^3)^{1/5} \approx 4.8$, e.g., from 2.1 to 10.0 °K. In order to obtain the near perfect fit (shown in Fig. 2) of the calculated resistivity to Eq. (21), particularly at the low-temperature end, it was necessary to subtract from $\rho(T)$ a ρ_0 that differed from the "true" ρ_0 by less than 0.1%. Even better fits to Eq. (21) are obtained for equally large ranges of P, where the magnitude of the individual P's are smaller, e.g., $10^3 \leq P \approx 0.4$. This can be thought of as corresponding to the same temperature range as $10^4 \leq P \leq 4$ but for a sample with a lower residual resistivity. In general, the magnitude of A increases with increasing values of the residual resistivity but much more slowly than in the case of true enhancement of electron-electron scattering processes, e.g., Ni in Pd.[2]

In summary, the theory of Klemens and Jackson, whose basic ideas have been experimentally verified, has been extended in a rigorous manner to include impurity scattering. The results deviate seriously from Matthiessen's rule in the temperature range where the residual resistivity dominates. The resulting resistivity can be fitted very well to Eq. (21). Great care, therefore, must be taken in attributing the appearance of an apparent T^2 term in the low-temperature resistivity in simple metals to the presence of electron-electron scattering.

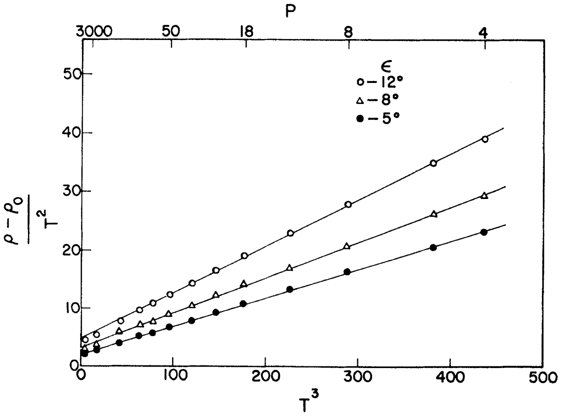

FIG. 2. The temperature-dependent resistivity divided by T^2 versus T^3 for fcc crystals with $\epsilon = 5°$, 8°, and 12°. The units of temperature and resistivity are arbitrary, depending only on some arbitrary choice of T (at some given P) and ρ_0. The magnitudes of P which are not arbitrary are given on the upper abscissa. Straight lines have been drawn through the points.

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Electrical Resistivity, Thermoelectric Power, and X-Ray Interference Function of Amorphous Ni-Pt-P Alloys*

Ashok K. Sinha
W. M. Keck Laboratory of Engineering Materials, California Institute of Technology, Pasadena, California 91109

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Amorphous alloys having the composition $(\mathrm{Ni}_x\mathrm{Pt}_{1-x})_{0.75}\mathrm{P}_{0.25}$, where $0.20 \le x \le 0.60$, were obtained by rapid quenching from the liquid state. X-ray diffraction measurements indicate a high degree of structural disorder in these alloys and an atomic configuration closely similar to that in liquid metals. At room temperature, the electrical resistivity ρ of these alloys lies between 160–185 $\mu\Omega$ cm, and the absolute thermoelectric power S between 1.9–2.5 $\mu V/{}^\circ K$. On increasing the temperature from 4.2 to 420 °K, up to which the amorphous alloys are stable, the resistivity of the alloy with $x=0.20$ decreases by about 2%; the value of $d\rho/dT$ progressively increases with increasing Ni content, becoming positive at $0.50<x<0.60$. In the range 80–300 °K, the dS/dT of all alloys lies between $5\text{–}8\times10^{-3}$ $\mu V\mathrm{deg}^{-2}$. The electrical behavior of these alloys may be treated in terms of electron scattering in disordered structures assuming the nearly free-electron model, in a manner analogous to Ziman's theory of electronic transport in liquid metals. The $d\rho/dT$ of these alloys is then qualitatively explained in terms of the temperature and composition dependence of the x-ray interference function $a(K)$, assuming an average number of ~ 1.3 conduction electrons per atom in these alloys. For the alloy with $x=0.20$, this implies a Fermi energy of 6.9 eV which corresponds to the position of the first peak in $a(K)$. The thermoelectric-power results lead to the conclusion that the "average" form factor for scattering of electrons decreases with increasing Ni/Pt ratio.

INTRODUCTION

In recent years there has been a continuing interest in the study of the behavior of electrons in disordered structures, especially as observed in the case of glassy semiconductors and liquid metals.[1, 2] It has been shown that the electrical transport properties of metallic liquids are greatly influenced by the "pair" correlation present in their structures. There is also a growing class of glassy metallic materials which may be obtained by rapid quenching from the liquid state.[3] The bulk transport properties of such materials would be expected to be predominantly influenced by their intrinsic structural disorder, somewhat analogous to the case of liquid metals. No experimental investigation concerning the structural scattering of electrons in such amorphous metallic alloys appears to have been reported as yet.

In the present paper we report the results on measurements of electrical resistivity and thermoelectric power as a function of temperature for a series of new amorphous alloys having the composition $(\mathrm{Ni}_x\mathrm{Pt}_{1-x})_{0.75}\mathrm{P}_{0.25}$ $(0.20 \le \chi \le 0.60)$. These alloys were obtained by rapid chilling from the liquid state. The structure parameter in terms of which these transport properties may be discussed is the x-ray interference function, and this was determined for two alloys having values of x equal to 0.20 and 0.50. A detailed radial distribution function study has led to the conclusion that these alloys possess a higher degree of disorder than that known in any of the existing glassy metallic alloys within their category.[4]

EXPERIMENTAL

The starting materials were metal powders of 99.99+ purity and reagent grade red phosphorus powder. The properly mixed alloy constituents