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

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Amorphous alloys having the composition $(\mathrm{Ni}_x\mathrm{Pt}_{1-x})_{0.75}\mathrm{P}_{0.25}$, where $0.20 \leq x \leq 0.60$, 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 $\mu V/°K$. 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 $5-8\times10^{-3}$ $\mu V\mathrm{deg}^{-2}$. 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 $(\mathrm{Ni}_x\mathrm{Pt}_{1-x})_{0.75}\mathrm{P}_{0.25}$ $(0.20 \leq \chi \leq 0.60)$. 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

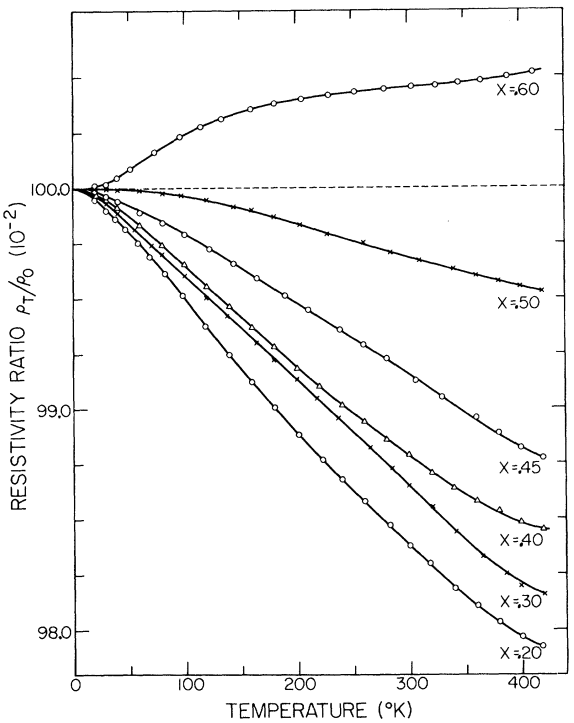

FIG. 1. Electrical resistivity versus temperature of amorphous $(Ni_xPt_{1-x})_{0.75}P_{0.25}$ alloys normalized with respect to the resistivity (extrapolated) at 0 °K. Not all the data points have been shown.

were compacted into briquettes under 50 000 ψ pressure, and subjected to "reactive sintering" by slowly heating them in evacuated fused silica capsules up to 500°C and holding there for two days. The sintered alloys were then melted by slowly raising their temperature up to 650°C. The total weight loss during alloy preparation was always less than 2% by weight, and for this reason the alloys will be referred to by their nominal (unanalyzed) compositions. Rapid quenching from the melt was effected using the piston and anvil technique[5] in which the quenching rates approach 10^6°C/sec.[6] The quenched foils were about 35 μ thick and 2.5 cm in diameter. Owing to the inexact reproducibility of the quenching conditions, it was necessary to always check the structure of the quenched foils. This was done by taking their x-ray diffraction pattern in the region of the first broad band. A step scan diffractometer using Cu$K\alpha$ radiation was employed. The presence of any microcrystals could be detected by the appearance of a few Bragg-like maxima superimposed upon the smooth broad band characteristic of a glassy structure.

For electrical measurements, strips were cut from the quenched foils. The electrical resistivity was measured in the temperature range 4.2–700°K using the standard four-probe method. The Pt leads were spot welded, and emf was measured using a Leeds and Northrup-type K-5 potentiometer. The relative values of resistance ratios are estimated to be accurate within ±0.01%. The thermal emf's were also potentiometrically measured. For temperature measurements, Cu-constantan thermocouples were spot welded on to the specimen. The over-all uncertainty in the thermoelectric-power measurements is estimated to be ±1%.

For determination of the interference function, the entire x-ray diffraction pattern was recorded in the range $12° \leq 2\theta \leq 180°$ using Mo$K\alpha$ radiation. A GE diffractometer fitted with a curved crystal diffracted beam monochromator, a pulse-height analyzer and a scintillation counter were employed. The specimen consisted of four foils glued together with Duco cement on a bakelite substrate. Such a specimen may be considered to be infinitely thick for the purpose of x-ray absorption corrections.[4] After making the corrections for polarization and background, the observed coherent intensities (in arbitrary units) were converted into those in electron units per atom (I_c^{eu}) using the high-angle method.[7]

RESULTS

Figure 1 shows the temperature dependence of resistivity of the present alloys. The resistivity values have been normalized with respect to the extrapolated resistivity ρ_0 at 0 °K.[8] The interesting feature of these results is that $d\rho/dT$ is negative for Pt-rich alloys. With increasing Ni content,

TABLE I. Electrical resistivity (ρ) and absolute thermoelectric power (S) of amorphous Ni-Pt-P alloys.

Alloy composition	$\rho_{300\,°K}$ ($\mu\Omega$ cm ±15)	$\frac{1}{\rho_0}\left(\frac{\partial\rho}{\partial T}\right)_{T=300\,°K}$ (10^{-5} °K^{-1})	$S_{300\,°K}$ (μV °K^{-1})	$S_{90\,°K}$ (μV °K^{-1})
$(Ni_{0.20}Pt_{0.80})_{0.75}P_{0.25}$	185	−4.9	2.16	0.90
$(Ni_{0.30}Pt_{0.70})_{0.75}P_{0.25}$	183	−4.7	2.28	0.86
$(Ni_{0.40}Pt_{0.60})_{0.75}P_{0.25}$	166	−3.9	1.97	0.85
$(Ni_{0.45}Pt_{0.55})_{0.75}P_{0.25}$	173	−3.4	1.90	0.91
$(Ni_{0.50}Pt_{0.50})_{0.75}P_{0.25}$	164	−1.4	2.54	0.90
$(Ni_{0.60}Pt_{0.40})_{0.75}P_{0.25}$	170	+0.4	2.50	0.87

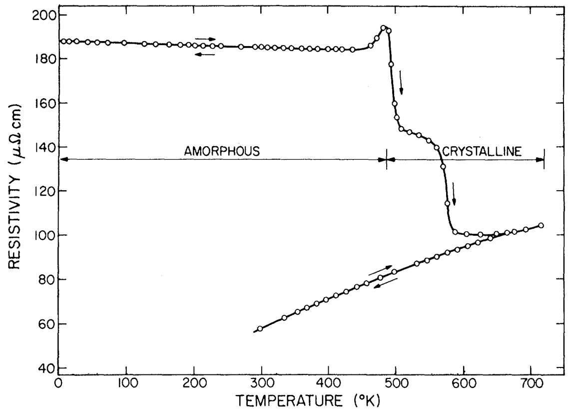

FIG. 2. Temperature dependence of electrical resistivity of an amorphous and crystallized $(Ni_xPt_{1-x})_{0.75}P_{0.25}$ alloy ($x = 0.20$).

$d\rho/dT$ becomes less negative and changes sign from negative to positive between $x = 0.50$ and $x = 0.60$. The room-temperature resistivities of all alloys are similar, and have values between 160 and 185 $\mu\Omega$ cm (Table I).

On heating slowly at a rate of 1.5 °K per min these alloys progressively transform into the equilibrium crystalline phases at temperatures above about 425 °K (Fig. 2). The onset of devitrification is marked by a small increase in the resistance owing to the nucleation of microcrystals,[9] and this is followed by a rapid decrease until the recrystallization is complete at ~650 °K. In the temperature range 650 – 300 °K, the crystallized alloy has a relatively large and positive $d\rho/dT$, and a room-temperature resistivity which is about 30% of the value for the amorphous phase.

The thermoelectric-power measurements were made relative to copper and the results were then converted into absolute units using the data of Cusack and Kendall.[10] All the alloys show a linear temperature dependence of thermoelectric power; the S-versus-T curves for two typical alloys are shown in Fig. 3. The values of S at 300 and 90 °K are listed in Table I.

The interference function $a(K)$ was obtained from the x-ray diffraction measurements. With a single x-ray diffraction experiment on each ternary alloy, it is not possible to obtain the six partial interference functions corresponding to each of the "pair" correlations present. The presently determined $a(K)$ shown in Fig. 4 corresponds to the convolution broadened weighted average atomic radial distribution function (RDF) $4\pi r^2 p(r)$ for the alloy as a whole.[4, 11] The following equations for $a(K)$ per atom may then be written:

$$K[a(K)-1] = \int_0^\infty 4\pi r[p(r) - p_0] \sin Kr\, dr , \qquad (1)$$

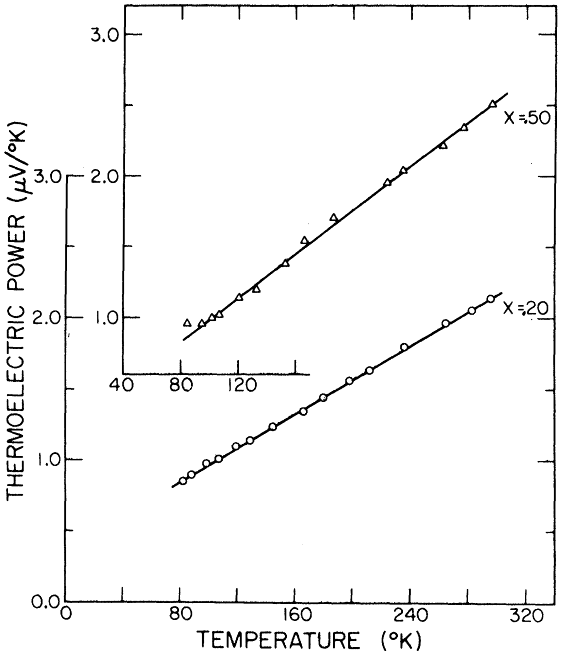

FIG. 3. Absolute thermoelectric power versus temperature for two $(Ni_xPt_{1-x})_{0.75}P_{0.25}$ alloys.

$$a(K) = 1 + (I_c^{eu} - \textstyle\sum_m c_m f_m^2) / (\sum_m c_m f_m)^2 , \qquad (2)$$

where K and r refer to the modulus of the diffraction vector in Fourier space and the position vector in the real space, respectively; p_0 is the average atomic density; $m, n, \ldots$ refer to the alloying elements Ni, Pt, ...; c is the atomic fraction; f is the modulus of the x-ray form factor corrected for anomalous dispersion. In the case of elastic scattering from state $\vec{k}_1$ to $\vec{k}_2$, we have

$$K = 2k \sin\theta = (4\pi/\lambda) \sin\theta , \qquad (3)$$

where, k is the wave number, λ is the wave length, and 2θ is the diffraction angle. As may be seen in Fig. 4, the function $a(K)$ has fairly well-defined peaks which get progressively smaller with increasing values of K and eventually degenerate into very weak modulations about the horizontal line, $a(K) = 1$, which represents a completely uncorrelated structure.

DISCUSSION

The electrical resistivities of the present alloys are similar in magnitude to those observed for liquid transition metals. The negative value of $d\rho/dT$ for Pt-rich alloys is particularly interesting in view of the fact that there is no experimental indication of localized magnetic moments.[12] In certain amorphous Pd-based alloys localized moments are known to give rise to a negative resistivity

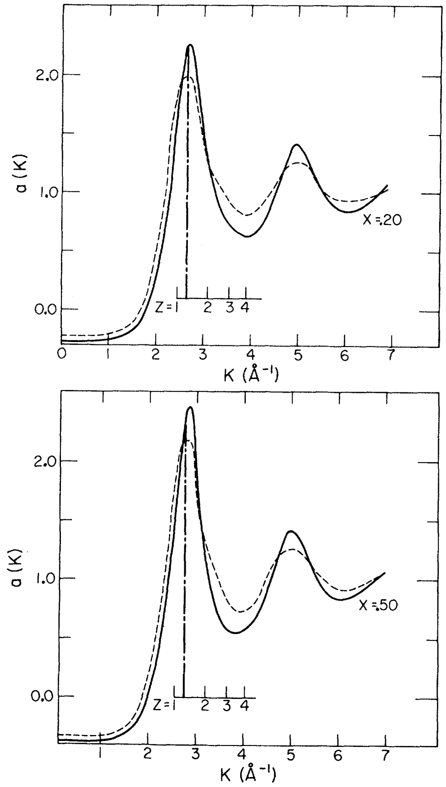

FIG. 4. Experimentally determined x-ray interference function versus $K(=4\pi/\lambda\,\mathrm{Sin}\theta)$ for two amorphous $(\mathrm{Ni}_x\mathrm{Pt}_{1-x})_{0.75}\mathrm{P}_{0.25}$ alloys (thick curves). Dashed curves show (schematically, following Ref. 23) $a(K)$ at higher temperature. The value of $2k_F$ for the number of conduction electrons per atom, $Z=1, 2, \ldots,$ is also shown. The vertical line represents the present alloy compositions with $Z=1.3$.

contribution through s-d spin-exchange interaction.[13] In the following discussion, an interpretation will be provided of the electrical behavior of present alloys in terms of the theories of electron scattering in disordered structures. It appears desirable therefore to briefly summarize at this point, the experimental evidence indicating a high degree of structural disorder in these alloys.

The shapes of the present $a(K)$ as well as the RDF curves are closely similar to those observed for liquid metals. Particularly noteworthy is the fact that unlike the case with all previously studied amorphous metallic alloys,[14-17] there is no shoulder on the high-angle side of the second peak in $a(K)$ (Fig. 4). Consequently, the RDF does not have a double peak beyond the first maximum.[4] Also, the ratio of the second to first near neighbor atomic distances r_2/r_1 ($=1.86$) lies within the range observed for liquid metals,[4] and is significantly higher than the values (1.6–1.8) observed for the previously studied amorphous films.[14]

The effect of structural disorder on the electronic properties is best considered through the use of the interference function $a(K)$. The peaks in $a(K)$ curve correspond to ripples in the E-k curve which are similar to the sharper Brillouin zone effects in crystalline solids.[18] In theories of elastic scattering of x rays[19] as well as of electrons[20] in disordered structures, particularly by liquids in the latter case, the importance of $a(K)$ has been recognized for some time. More recently Ziman and his collaborators[21, 22] have shown that metallic conduction in simple liquids may be treated as a case of weak scattering of electrons in the nearly free-electron (NFE) model. The matrix element responsible for scattering is $\langle \vec{k}_1 | U | \vec{k}_2 \rangle$, where U is a nonlocalized pseudopotential operator, and $\vec{k}_1$ and $\vec{k}_2$ are two states on the Fermi surface. The differential scattering cross section may then be obtained using the Born approximation. The equation for the relaxation time τ which appears in the solution of Boltzmann transport equation may be written in the following form:

$$\frac{1}{\tau}=\frac{mk_F}{\pi\hbar^3 V}\int_0^{\pi/2} |u(K)|^2\, a(K) \sin 2\theta\, (1-\cos 2\theta)\, d\theta \;, \tag{4}$$

where m is the electronic mass, $\vec{k}_F$ is the wave vector on Fermi surface, and V is the atomic volume. The value of $1/\tau$ depends upon the square of the Fourier component of the pseudopotential $|u(K)|^2$, which acts like a form factor for electron scattering and upon the interference function $a(K)$, which is equivalent to a structure factor. Using Eq. (3), the above equation may be rewritten as follows:

$$\frac{1}{\tau}=(2mk_F/\pi\hbar^3 V)\langle |u(K)|^2\, a(K)\rangle \,, \tag{5}$$

where $$\langle g(K)\rangle = (1/4k_F^4)\int_0^{2k_F} g(K)\, K^3\, dK \;. \tag{6}$$

Substituting Eq. (4) into the usual expression for electrical resistivity, $\rho=(Vm/Ze^2)\,1/\tau$, we get

$$\rho=(2m^2k_F/\pi Z e^2\hbar^3)\;\langle\, |u(K)|^2 a(K)\rangle \,, \tag{7}$$

where e is the electronic charge and Z is the average number of conduction electrons per atom.

Recently the pseudopotential approach has been extended to transition metals in which the partial filling of the d band may be neglected.[23] Such a situation probably exists in the present alloys owing to the possibility of electron transfer from phosphorus into the vacant states of Ni and Pt. $\langle |u(K)|^2\rangle$ in such a case would then include the sum of a transition metal pseudopotential form factor and the form factor for a screened hybridization term.[24] However, we may assume $\langle |u(K)|^2\rangle$ to be a function of K only,[21] so that in discussing the temperature variation of resistivity of amorphous alloys, a detailed knowledge of the individual pseudopotentials is not necessary.

It is then possible to qualitatively explain the negative temperature coefficient of resistivity of amorphous $(\mathrm{Ni}_{0.20}\mathrm{Pt}_{0.80})_{0.75}\mathrm{P}_{0.25}$ alloy by making the following two assumptions: (a) $2k_F$ for this alloy is approximately equal to the value of K corresponding to the first peak in $a(K)$. (b) The first peak of $a(K)$ broadens and decreases in height with increasing temperature. Such a temperature dependence of $a(K)$ was experimentally established for a liquid Cu-Sn alloy by Halder *et al.*[25] Here $2k_F$ falls on the position K_1 of the first peak of x ray $a(K)$ and the observed decrease in peak height of about 18% for a 360°C temperature increase is adequate to explain the negative $d\rho/dT$ of this alloy. The neutron diffraction work of Wingfield *et al.*[26] on liquid zinc also indicates a general tendency towards a temperature-induced broadening in the $a(K)$ peak; however, the change in the peak height was found to be too small to account for a temperature coefficient of resistivity (TCR) of about $-130\,\mathrm{ppm}/{}^\circ\mathrm{C}$ for liquid zinc. It should be pointed out that in the case of a solid amorphous alloy like $(\mathrm{Ni}_{0.20}\mathrm{Pt}_{0.80})_{0.75}\mathrm{P}_{0.25}$, a very small decrease in $a(K)$ peak height would be necessary to explain its TCR of $-49\,\mathrm{ppm}/{}^\circ\mathrm{C}$. The situation is schematically illustrated in Fig. 4. The position of the first peak in $a(K)$ corresponds to a Fermi energy $E_F \approx 6.9$ eV and an average number of conduction electrons per atom $Z \approx 1.3$. These quantities are related by the NFE relation $Z\rho_0 = k_F^3/3\pi^2$ where ρ_0 is equal to the experimentally determined density of 0.063 atoms/Å^3. An increase in the Ni content of the alloy causes a shift in the first peak of $a(K)$ towards higher K values.[27] This would have the effect of gradually moving $2k_F$, corresponding to the same value of Z, towards a region where $a(K)$ increases with increasing temperature. Such a region, of course, corresponds to a positive $d\rho/dT$. The presently estimated number of 1.3 *s* electrons per atom for amorphous Ni-Pt-P alloys is significantly higher than that (0.5 – 0.6) known to exist in Pt or Ni. Such a high number would suggest that the phosphorus atoms in such alloys exist in a highly ionized state, and contribute their valence electrons into the conduction band.

Ziman's theory can also be used to obtain information on another derivative of resistivity, namely, $d\rho/dE$. Thus, for the diffusion part of thermoelectric power, S, we have the following expression:

$$S = \frac{\pi^2 k_B^2 T}{3|e|}\left(\frac{\partial \ln\rho(E)}{\partial E}\right)_{E=E_F}, \qquad (8)$$

where k_B is the Boltzmann constant. Upon substituting Eq. (7) into above, we get

$$\frac{2|u(2k_F)|^2}{\langle |u(K)|^2\rangle}\,\frac{a(2k_F)}{\langle a(K)\rangle} = 3 + \frac{3|e|E_F}{\pi^2 k_B^2}\,\frac{S}{T}\,. \qquad (9)$$

For the alloy $(\mathrm{Ni}_{0.20}\mathrm{Pt}_{0.80})_{0.75}\mathrm{P}_{0.25}$, the right-hand side of Eq. (9) was evaluated using the experimentally determined value of $S_{300\,^\circ\mathrm{K}}$ and $E_F = 6.9_4$ eV, and is found to be equal to 5.0_4. $a(2k_F)/\langle a(K)\rangle$ can be evaluated using the data in Fig. 4 and is found to be $\sim 4.2_3$. This gives a value of 0.6 for $|u(2k_F)|^2/\langle |u(K)|^2\rangle$. In view of the simplifications made in the theory, such numerical values must be considered as indications only of the order of magnitude of the corresponding qualities. However, the fact that $|u(2k_F)|^2/\langle |u(K)|^2\rangle$ appears to be less than 1 would suggest that in this alloy $u(K)$ is decreasing with increasing values of K. If we assume the present alloys to be associated with a constant value of Z, it means that the "average" form factor for electron scattering $u(K)$ is decreasing with increasing Ni/Pt ratio in the alloy.

CONCLUSIONS

(1) Ternary alloys having the composition $(\mathrm{Ni}_x\mathrm{Pt}_{1-x})_{0.75}\mathrm{P}_{0.25}$ where $0.20 \le x \le 0.60$, may be obtained in the glassy state by rapid liquid quenching technique. The atomic configuration in these alloys closely resembles that found in liquid metals. Their electrical properties can be interpreted in terms of electron scattering in a disordered structure, assuming a spherical Fermi surface (NFE model).

(2) The negative $d\rho/dT$ for amorphous $(\mathrm{Ni}_{0.20}\mathrm{Pt}_{0.80})_{0.75}\mathrm{P}_{0.25}$ appears to be due to its high Fermi energy of about 6.9 eV; this corresponds to the position of the first peak in its x-ray interference function $a(K)$ at which $a(K)$ decreases with increasing temperature. Assuming a constant number of conduction electrons per atom of ~ 1.3, the progressive change in $d\rho/dT$ towards positive values on increasing x may be qualitatively explained by the shift in the peak of $a(K)$ towards higher K values; this causes $K(=2k_F)$ corresponding to the alloy composition move into a region where $a(K)$ increases with increasing temperature.

(3) On the basis of the observed linear temperature dependence of the positive thermoelectric power of these alloys, it would appear that the "average" form factor for electron scattering decreases with increasing Ni content.

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Ultrasonic Attenuation Due to Electron-Phonon Interaction in Potassium

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The electron-phonon contribution to the ultrasonic attenuation and the electrical resistivity of potassium at low temperatures is calculated using a number of different pseudopotentials. Serious discrepancies are found between the calculated values and the experimental data reported by Natale and Rudnick.