Electronic transport in amorphous Ni$_{1-x}$P$_x$ alloys

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We have measured the resistivity $\rho(T)$ and the thermoelectric power $Q(T)$ in the metallic glass system Ni$_{1-x}$P$_x$, with $0.143 \leq x \leq 0.243$, prepared by chemical deposition and melt quenching. We find that as x increases, ρ increases, α [$=(1/\rho)d\rho/dT$] decreases and becomes negative, and Q increases and becomes positive near $x=0.18$. These results agree with the Mooij correlation and with a correlation of positive Q with high ρ and negative Q with low ρ, which is also seen in many nonmagnetic metallic glasses. Thus the Ni$_{1-x}$P$_x$ system spans the range of behavior seen in many different metallic glasses. For $x \leq 0.175$, $\rho(T)$ and $Q(T)$ are very similar to those seen in iron-based ferromagnetic glasses. The only clear difference between the transport properties of samples prepared by melt quenching and chemical deposition is in the temperature dependence of α. We compare our results with several theories for electron scattering.

I. INTRODUCTION

Electrical transport in metallic glasses has attracted considerable attention because of features not found in ordinary crystalline metals. For example, some glasses consisting of alloys of nonmagnetic elements, such as Be$_{0.4}$Ti$_{0.5}$Zr$_{0.1}$ (Ref. 1), Cu$_{0.5}$Zr$_{0.5}$, (Ref. 2), or Ca$_{0.8}$Al$_{0.2}$ (Ref. 3) have large resistivities, ρ, which decrease slowly and monotonically with increasing temperature. The larger the magnitude of ρ the more negative is the temperature coefficient of resistivity, $\alpha=(1/\rho)d\rho/dT$. In contrast, resistivities of normal crystalline metals are usually smaller and increase rapidly with increasing temperature. This dependence in metallic glasses of α on the magnitude of ρ is an example of a general trend discovered by Mooij in 1973.[4] By looking at a variety of metallic systems Mooij found that α was lower in systems of higher ρ; in particular, most systems with $\rho < 150$ $\mu\Omega$ cm have a positive α and those with $\rho > 150$ $\mu\Omega$ cm have a negative α.

Another group of metallic glasses, containing ferro- or antiferromagnetic elements, shows a second kind of unusual behavior. Alloys such as Fe$_{0.8}$B$_{0.2}$ or Fe$_{0.32}$Ni$_{0.36}$Cr$_{0.14}$P$_{0.12}$B$_{0.06}$ have positive values of α at room temperature, however, α changes sign below room temperature where the resistivity has its minimum value.[5] This is reminiscent of the Kondo effect which occurs in nonferromagnetic crystalline metals with a dilute amount of ferromagnetic impurities.[6] However, this effect is not seen in crystalline ferromagnetic metals in which the spins of the lattice cannot freely flip to scatter electrons. Thus it is unclear whether the Kondo effect can explain the minimum in ρ in ferromagnetic metallic glasses.

Another transport property in metallic glasses which has interesting behavior is the thermoelectric power $Q(T)$. In the nonmagnetic metallic glasses with $\alpha < 0$, such as Be$_{0.4}$Ti$_{0.5}$Zr$_{0.1}$ (Ref. 1), Cu$_{0.5}$Zr$_{0.5}$ (Ref. 2), or Ca$_{0.8}$Al$_{0.2}$ (Ref. 7), $Q(T)$ is small and close to linear over a wide range of temperatures. In crystalline metals, one has never observed linear thermoelectric powers even though simple theories of the thermoelectric power predict a linear dependence of Q on T.

A correlation is seen between the sign of the thermoelectric power and the sign of the temperature coefficient of resistivity (or the magnitude of ρ) in nonmagnetic metallic glasses. The glasses mentioned above all have $\rho > 150$ $\mu\Omega$ cm, $\alpha < 0$, and positive Q. In Pd$_{0.775}$Cu$_{0.06}$Si$_{0.165}$, which has $\rho=85$ $\mu\Omega$ cm and a positive α, Q is negative.[8] The correlation of positive Q with high ρ and negative Q with low ρ is very similar to the Mooij correlation exhibited by these same glasses. There has been little systematic investigation of the thermoelectric power, however. The purpose of this paper is to report a measurement of $Q(T)$ in a single metallic glass sys-

tem, $Ni_{1-x}P_x$. By varying the composition in this alloy the resistivity can be varied from 100 to 170 $\mu\Omega$ cm (Ref. 9) (thus spanning the value 150 $\mu\Omega$ cm, the resistivity which appears as the crossover point in the Mooij correlation). At the same time, α is positive at the lowest resistivities and decreases and becomes negative as the resistivity is increased. Therefore, we can observe Q in a single system which spans the range of behavior of the resistivity in nonmagnetic metallic glasses.

In Sec. II we discuss several models for scattering mechanisms which can lead to a decreasing resistivity with increasing temperature and their predictions for the thermoelectric power. Section III deals with experimental technique. The results are presented in Sec. IV, and a discussion follows in Sec. V.

II. BACKGROUND

Several scattering mechanisms have been proposed to explain the negative values of α found at all temperatures in metallic glasses: (a) scattering from structural disorder, the Ziman theory, (b) Mott s-d scattering, (c) scattering from structural two-level systems, and (d) theories of incipient localization.

(a) In the Ziman theory the electrons scatter from the disordered arrangement of atomic potentials.[10] This can lead to both an increasing or decreasing resistivity with increasing temperature depending on the relative magnitudes of the Fermi wave vector and the position of the first peak in the static structure factor. Originally proposed for simple liquid metals, the Ziman theory has been extended to include liquid–transition-metal alloys.[11] Because of the similarity of the disordered structures of metallic glasses and liquid metals the Ziman theory has subsequently been applied to metallic glasses.[1,12]

As in the case of transition-metal liquids, in a glass containing a single transition metal the resistivity is dominated by the d-wave phase shift and backward scattering. By assuming independent atomic vibrations, the resistivity as a function of temperature for such a glass is approximately[11,13]

$$\rho(T)=\frac{30\pi^3\hbar^3}{me^2k_F^2E_F\Omega}\sin^2[\eta_2(E_F)]$$

$$\times\{1+[S_0(2k_F)-1]e^{-2[W(T)-W(0)]}\}\ .$$

$$(1)$$

Here k_F and E_F are the Fermi wave vector and energy, Ω is the atomic volume, $\eta_2(E_F)$ is the d-wave phase shift, $S_0(k)$ is the static structure factor at temperature $T=0$, and $W(T)$ is the Debye-Waller exponent at temperature T. $W(T)$ varies as T^2

when $T\ll\Theta_D$ (Θ_D is the Debye temperature) and as T when $T>\Theta_D$. The condition for a negative temperature coefficient of resistivity in this theory is when $2k_F\approx k_p$ (where k_p is the position of the first peak in the structure factor) in which case $S_0(2k_F)>1$. The measured change in the peak of the structure factor with temperature approximately accounts for the change seen in the resistivity in the case of $Nb_{1-x}Ni_x$, $x=0.4$, 0.5, and 0.6.[14] The Ziman theory also predicts a positive temperature coefficient of resistivity when $S_0(2k_F)<1$. This will happen if $2k_F$ is far from a peak in the structure factor. With more than one transition metal a similar analysis holds except that the phase shift for each transition-metal component and the partial structure factors must be used.[15]

(b) Mott s-d scattering assumes that the electrons near the Fermi surface can be divided into two groups with very different mobilities.[16] The more mobile s or p carriers are strongly scattered into the less mobile d holes at the Fermi surface. The resistivity which is dominated by this scattering process is proportional to the number of such holes, i.e., to $N_d(E_F)$, the density of d states at the Fermi energy. In metals with partially full d bands $N_d(E)$ changes rapidly at E_F and ρ decreases with increasing temperature due to the thermal broadening of the electron distribution and the motion of the chemical potential. With the use of a simplified model where $\rho(E)\propto N_d(E)$ and the density of states does not change with temperature, the resistivity in this theory is[17]

$$\rho(T)=\rho(0)\left\{1-\frac{\pi^2}{6}(k_BT)^2\left[3\left[\frac{1}{N_d}\frac{dN_d}{dE}\right]^2\right.\right.$$

$$\left.\left.-\frac{1}{N_d}\frac{d^2N_d}{dE^2}\right]_{E_F}\right\}\ .$$

$$(2)$$

If we assume for simplicity that the d band is nearly full and that $N_d(E)=C(E_0-E)^{1/2}$ (with $E_F<E_0$), then

$$\rho(T)=\rho(0)\left[1-\frac{\pi^2}{6}(k_BT)^2(E_0-E_F)^{-2}\right]\ .\quad(3)$$

Brouers and Brauwers[18] have considered the possibility that $N_d(E)$ does depend on temperature and found that there is a significant smearing out of $N_d(E)$ with increasing temperature. Aside from the effects of the thermal broadening of the electron distribution, this modification of $N_d(E)$ and the shift in E_F which is caused by it lead to a contribution to the resistivity which can either increase or decrease with temperature in a manner consistent with the

Mooij correlation. When E_F is close to a maximum in $N_d(E)$, the resistivity is high and thermal smearing causes $N_d(E_F)$ and ρ to decrease with increasing temperature. When E_F is far from a maximum in $N_d(E)$, the resistivity is low and the smearing causes $N_d(E_F)$ and ρ to increase with increasing temperature. A more complete theory of s-d scattering must take into account both the thermal broadening of the electron distribution and the thermal smearing of $N_d(E)$. For our purposes we only need to note that both effects produce a negative α when E_F is close to a maximum in $N_d(E)$ and that α becomes more positive as $[d(\ln N_d)/dE]^2_{E_F}$ and $N_d(E_F)$ are decreased.

Mott has stressed that the model of s-d scattering can only hold when there is a significant difference in the mobilities of the s and d electrons.[16] Thus this model may break down for very-high-resistivity materials where all mobilities are very low such as in many metallic glasses.

(c) Two-level tunneling systems similar to those proposed by Anderson et al.[19] and Phillips[20] in insulating glasses have also been used to try to explain some of the unusual electronic transport behavior in metallic glasses. The interaction between these systems and the conduction electrons produces a contribution to the resistivity which increases with decreasing temperature analogous to the Kondo effect. Cochrane et al.[21] used this model to explain the low-temperature minimum found in the resistivity of some metallic glasses, as mentioned in Sec. I, which resembles the minimum seen in crystalline Kondo systems. They calculated a term in ρ of the form $-C\ln(T^2+\Delta^2)$, where $2k_B\Delta$ is the energy splitting between the tunneling states. Tsuei[22] has suggested that this mechanism may be responsible for producing a negative α at all temperatures in high-resistivity metals. He found a better fit to $\rho(T)$ data in these metals with the form $\rho(T)=\rho(0) -C\ln(T^2+\Delta^2)$ than with the Ziman theory's linear decrease in resistivity with temperature at high temperatures.

(d) The theory of localization has been used by Jonson and Girvin[23] and by Imry[24] to explain the Mooij correlation on very general grounds. These theories also have the advantage that they begin in the limit of strong scattering, i.e., assuming the electronic mean free path is very short as it is in metallic glasses. The scattering mechanisms described above, on the other hand, are essentially weak scattering theories and may not be applicable in the strong scattering limit. According to Jonson and Girvin, in low-resistivity metals phonons scatter electrons and the resistivity increases with increasing temperature, whereas in high-resistivity materials the adiabatic approximation breaks down and the

phonon dynamics must be included in any picture of the conduction. The effect of the phonons when the electron mean free path is very short is to increase the mobility of the conduction electrons, even though the electrons are not localized. The Mooij correlation follows directly from this model. It is produced by a competition between the two effects of the phonons: increased scattering of electrons due to increased disorder and the increase in mobility of electrons due to inelastic processes.

Imry's arguments based on the scaling theory of localization lead to the same conclusions. As the temperature increases the electron-phonon inelastic mean free path decreases below the correlation length which determines the resistivity in the scaling theory in the extended phase. The resistivity is then proportional to the inelastic mean free path and, therefore, decreases with increasing temperature. Girvin and Jonson[25] have shown that Imry's arguments are essentially equivalent to their own.

Each of these theories is able to predict a decreasing resistivity with increasing temperature. In order to distinguish between them another transport property must be measured. The thermoelectric power is a good choice because it is very sensitive to electron scattering mechanisms. In simple theories it is related to the resistivity by the Mott formula

$$Q(T)=\frac{\pi^2 k_B^2 T}{3|e|}\left[\frac{d\ln\rho}{dE}\right]_{E_F}. \tag{4}$$

We briefly describe the predictions for Q for each of the scattering mechanisms discussed above.

(a) The thermoelectric power $Q(T)$ in the Ziman theory is[26]

$$Q(T)=-\frac{\pi^2 k_B^2 T}{3|e|E_F}(3-2q-\tfrac{1}{2}r), \tag{5}$$

where k_B is the Boltzmann constant, T is the absolute temperature, and

$$q=\frac{S(2k_F)|t(2k_F)|^2}{\int_0^{2k_F}dk[4(2k_F)^{-4}k^3 S(k)|t(k)|^2]}. \tag{6}$$

Here $t(k)$ is the t matrix describing the muffin-tin potential. The quantity r in the expression for $Q(T)$ is the energy dependence of the t matrix:

$$r=\frac{\int_0^{2k_F}dk[4(2k_F)^{-4}k^3 S(k)k_F d\,|t(k)|^2/dk_F]}{\int_0^{2k_F}dk[4(2k_F)^{-4}k^3 S(k)|t(k)|^2]}. \tag{7}$$

Note that $Q(T)$ is linear in temperature and can be either positive or negative depending on the magnitude of q. When $2k_F\approx k_p$ and $S(2k_F)$ is large then q

is large and the thermoelectric power is positive. The condition $2k_F \approx k_p$ is also what is required to make the temperature coefficient of resistivity negative and the resistivity large.

(b) In Mott s-d scattering the thermoelectric power can have either sign depending on the relative position of the Fermi level and the d-band density of states. Since $Q \propto [d(\ln\rho)/dE]_{E_F}$, and in Mott s-d scattering $\rho(E) \propto N_d(E)$, then $Q \propto [d(\ln N_d)/dE]_{E_F}$ in this model. For example, if the d band is nearly full this would predict a negative value of Q. The smearing out of $N_d(E)$ with temperature would not change the sign of Q but would affect its magnitude.

(c) In real Kondo systems anomalously large thermoelectric powers of up to 40 μV/K are seen near the Kondo temperature, the temperature characteristic of the electron-lattice spin interaction.[26] Scattering from structural two-level systems should produce an analogous effect. In metallic glasses the anomaly will not be as spectacular as in crystalline systems; the Nordheim-Gorter rule states that scattering mechanisms contribute to the thermoelectric power in proportion to their contribution to the resistivity. In the model of Cochrane *et al.* anomalies are expected at low temperatures in $Q(T)$. If the two-level system scattering is causing α to be negative at high temperatures as in the model of Tsuei, then $Q(T)$ should show anomalies at high temperatures as well.

(d) The localization theory of Girvin and Jonson makes no prediction for the sign of the thermoelectric power, but a linear $Q(T)$ is not inconsistent with their ideas.[27]

A common behavior of the thermoelectric power is seen in nonmagnetic metallic glasses with negative temperature coefficients of resistivity—$Q(T)$ is positive, nearly linear, and small. Negative α, large ρ ($\rho > 150$ $\mu\Omega$ cm), and positive and linear $Q(T)$ have been seen in $Be_{0.4}Ti_{0.5}Zr_{0.1}$ (Ref. 1), $Cu_{0.5}Zr_{0.5}$ (Ref. 2), $Nb_{0.5}Ni_{0.5}$ (Ref. 8), $Ca_{0.8}Al_{0.2}$ (Ref. 7), $(Ni_xPt_{1-x})_{0.75}P_{0.25}$ (for $x \leq 0.50$),[12] and $Ni_{0.76}P_{0.24}$.[28] Furthermore, $Pd_{0.775}Cu_{0.06}Si_{0.165}$, which has a lower resistivity than the above glasses ($\rho = 85$ $\mu\Omega$ cm), has a positive α and a negative (but nonlinear) $Q(T)$.[8] There is one system, $La_{1-x}Ga_x$, $x = 0.18$ and 0.22, in which the noted correlation between the sign of α and the sign of $Q(T)$ is not observed to hold.[29] In magnetic glasses much more complicated behavior is observed in the temperature dependence of Q.[2]

Most of the metallic glasses examined experimentally exhibit either high-resistivity behavior ($\rho > 150$ $\mu\Omega$ cm, $\alpha < 0$, $Q > 0$) or low-resistivity behavior ($\rho < 150$ $\mu\Omega$ cm, $\alpha > 0$, $Q < 0$). The alloys $Ni_{1-x}P_x$ provide a single-alloy system in which the resistivity can be smoothly varied from the low-resistivity re-

gime to the high-resistivity regime. At the same time, α is positive when ρ is low and decreases with increasing ρ. The thermoelectric power of one sample has been measured previously ($Ni_{0.76}P_{0.24}$) and a positive linear $Q(T)$ was found from 77 K to room temperature.[28]

Ni is a ferromagnetic element, however, and the metallic glasses $Ni_{1-x}P_x$ have interesting magnetic properties. In electrodeposited $Ni_{1-x}P_x$ Berrada *et al.*[30] found that samples with $x < 0.18$ were weak homogeneous ferromagnets. The Curie temperature and the saturation magnetization decreased rapidly with increasing x. Above $x = 0.18$ the glass is weakly paramagnetic with a susceptibility which decreases with x.[31] Minima in $\rho(T)$ were seen near 10 K similar to those seen in iron-based ferromagnetic metallic glasses.[30]

III. METHOD

Amorphous $Ni_{1-x}P_x$ samples were produced by three methods. Samples in the range $0.143 \leq x \leq 0.219$ were prepared by chemical reduction as described in Ref. 32. The solutions used contained $NiCl_2$ for $x = 0.143$ and $NiSO_4$ for the others. Rapid quenching of the melt, as described in Ref. 33, produced amorphous samples with $x = 0.181$, 0.202, and 0.213. The sample containing the highest concentration of phosphorus ($x = 0.243$) was made by an electrodeposition technique described in Ref. 34.

Electron microprobe analysis was used to measure the composition x (phosphorus atomic fraction) and homogeneity Δx (Table I). Absolute accuracy in x is ± 0.01; relative accuracy in x between samples is ± 0.005. Most of the samples were homogeneous but several showed microscopic inhomogeneities across their thickness (see Table I). All samples were examined for traces of crystallinity by x-ray diffraction; none was found. Analysis was performed for transition-metal impurities by atomic absorption spectrophotometry. The results are listed in Table I for the major magnetic impurity.

Thermal behavior was observed by differential scanning calorimetry (DSC) at a heating rate of 10 K/min. We found features similar to those described by Clements and Cantor[35] in chemically deposited samples in the range $x < 0.18$. The glass $Ni_{1-x}P_x$ with $x = 0.143$ produced three exothermic peaks (Fig. 1). The first, a broad and shallow peak from 500 to 560 K, corresponds to crystallization of the glass into a fcc solid solution of P in Ni according to Clements and Cantor. The peaks at high temperature, 605 and 620 K, were attributed to rearrangements from the disordered crystal to ordered crystalline phases. Schmidt *et al.*[36] attribute the low

TABLE I. $Ni_{1-x}P_x$ sample composition, preparation, and homogeneity.

Composition, x (at. fraction P)	Preparation method[a]	Inhomogeneity, Δx (at. fraction P)	Major magnetic impurity (at. ppm)	Temp. of first DSC Peak (K)
0.143	C $(NiCl_2)$	0.009	Fe (12)	530,605
0.172	C $(NiSO_4)$	<0.002	Fe (65)	603
0.175	C $(NiSO_4)$	0.007	Fe (63)	603
0.181	Q	<0.002	Fe (351)	600
0.196	C $(NiSO_4)$	<0.002	Fe (78)	620
0.202	Q	<0.002	Co (677)	630
0.213	Q	0.003	Fe (278)	612
0.219	C $(NiSO_4)$	<0.002	Fe (201)	610
0.243	E	0.006	Fe (581)	550

[a]C is chemically deposited, Q is melt quenched, and E is electrodeposited.

first peak to crystallization of Ni-rich clusters. The second and third peaks they attribute to the crystallization of the rest of the glass and to a separation of the crystalline phases, respectively. With one exception, the other samples showed one exothermic crystallization peak. The temperature at which the peak occurs, T_p, increases with x, from 603 K at $x=0.172$ to a maximum of 630 K at $x=0.202$. Above $x=0.202$, T_p decreases to 550 K at $x=0.243$ (Table I). The maximum in T_p at $x=0.202$ in our chemically deposited and melt quenched samples differs from the monotonic decrease in T_p with increasing x seen in electrodeposited $Ni_{1-x}P_x$ by Cote.[37] The sample with $x=0.175$ had two narrow peaks, at $T=603$ and 612 K, similar to the second and third peaks of the sample with $x=0.143$. The similarity with the double peaks found in $x=0.143$ may be due to the inhomogeneity of the $x=0.175$ sample (see Table I), causing islands of lower P concentration.

The resistivity of the samples at room temperature were measured by the four-probe resistance method. Length, mass, and density[36] measurements were used to determine the sample dimensions. The overall accuracy in the resistivity measurement is ±4–9%.

The temperature dependence of the resistivity was measured from $T=4$–295 K. The temperature was measured by a $AuFe_{0.07\%}$-Chromel thermocouple.
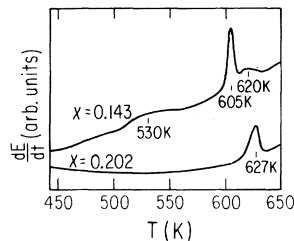
The accuracy in $\rho(T)/\rho(4.2$ K$)$ is $\pm5\times10^{-5}$.

The thermoelectric power was measured relative to $>99.999\%$-pure Pb from $T=4$–440 K. The integral method was used; one end of the sample was kept at a constant temperature while the other end was raised in temperature. The derivative of the voltage measured with respect to temperature gives the relative thermoelectric power. Below room temperature the absolute thermoelectric power of lead was taken from the scale of Roberts.[38] Above room temperature the scale of Cook, Laubitz, and Van der Meer[39] was used, adjusted to fit smoothly with the Roberts scale. The absolute thermoelectric power of the sample is obtained by subtracting the absolute thermoelectric power of the Pb standard from the measured relative thermoelectric power. Temperature was measured by copper-constantin thermocouples. A Keithley 180 nanovoltmeter was used to measure the sample vs Pb emf. Above $T=40$ K, the accuracy in $Q(T)$ is ±0.05 μV/K in the samples with $x\leq0.181$, and is ±0.10 μV/K in the samples with higher x.

IV. RESULTS

The absolute resistivity at room temperature $\rho(295$ K$)$ as a function of phosphorus atomic fraction x is shown in Fig. 2. The resistivity increases
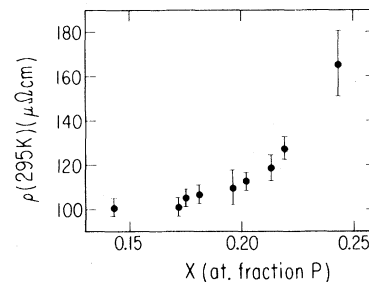

FIG. 1. Differential scanning calorimetry data taken at 10 K/min for $Ni_{1-x}P_x$ samples with $x=0.143$ and 0.202.

FIG. 2. Resistivity vs x, the atomic fraction of phosphorus.
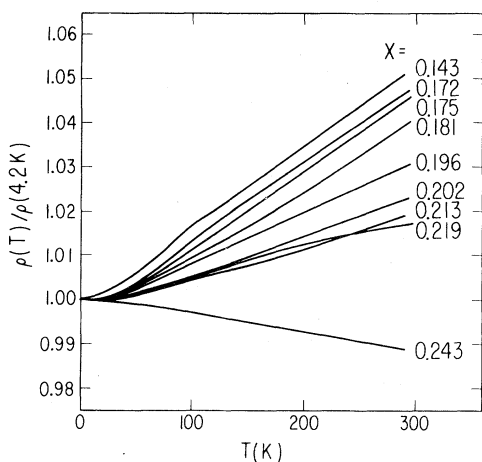
FIG. 3. Resistivity vs temperature for each $Ni_{1-x}P_x$ sample normalized to the sample's resistivity at 4.2 K.

monotonically as x is increased. Our measurements agree within experimental error with Cote's measurements on electrodepositied samples.[9]

The temperature dependence of the resistivity $\rho(T)/\rho(4.2$ K) for each sample is shown in Fig. 3. With increasing phosphorus content the temperature coefficient of resistivity $\alpha=(1/\rho)d\rho/dT$ decreases and changes sign, becoming negative near $x=0.23$. The decrease in α is monotonic in x except for the $x=0.175$ chemically deposited sample which has an anomalously large α below 100 K. One possible explanation is this sample's inhomogeneity (see Table I) although none of the other inhomogeneous samples show similar behavior.

The temperature dependence of α is also interesting. In Fig. 4, $\alpha(T)$ is shown for three pairs of samples with similar compositions, one chemically deposited (C) and one melt quenched (Q). For each chemically deposited sample, $\alpha(T)$ decreases above $T=80$ K, but for each melt quenched sample, $\alpha(T)$ increases above $T=80$ K.

The thermoelectric power $Q(T)$ is shown in Fig. 5 for each sample. The thermoelectric power increases with increasing phosphorus concentration and changes sign, becoming positive above $x=0.18$. The temperature dependence of Q is not linear over the entire measured temperature range for any of the samples. The samples with lowest x ($x=0.143$, 0.172) have negative $Q(T)$ with large positive curvatures which produce broad minima in $Q(T)$. This is also the ferromagnetic range of composition. Samples of intermediate-phosphorus content ($x=0.175$, 0.181) have small $Q(T)$ with both positive and negative curvatures in different temperature ranges. These compositions are near the transition from ferromagnetic to paramagnetic behavior. Samples with
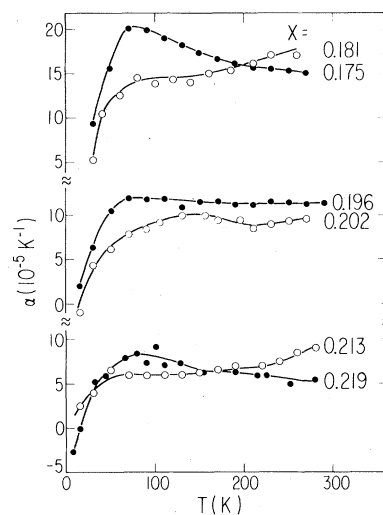

FIG. 4. Temperature coefficient of resistivity $\alpha=(1/\rho)d\rho/dT$ vs temperature for three pairs of $Ni_{1-x}P_x$ alloys with approximately the same x. The solid circles are for the chemically deposited samples and the open circles are for the melt quenched samples.

large x ($x=0.196$, 0.202, 0.213, 0.219, and 0.243) have positive $Q(T)$ with negative curvatures. In this range of compositions $Q(T)$ fits a power law in T, $Q(T)=CT^B$ with $B=0.7$, as seen in Fig. 6. The average curvatures are shown in Fig. 7.

V. DISCUSSION

By summarizing the results, as the phosphorus content is increased from $x=0.14$ to 0.24, the resistivity increases from 100 to 170 $\mu\Omega$ cm, the temperature coefficient of resistivity decreases smoothly
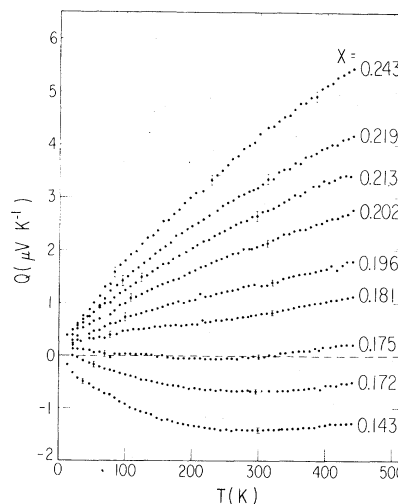

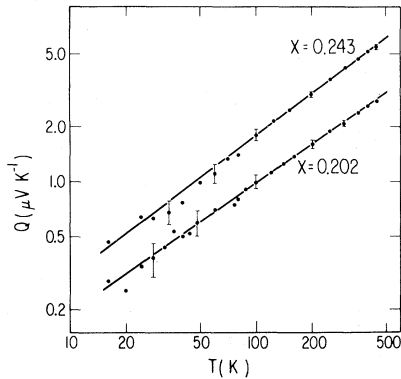FIG. 5. Thermoelectric power vs temperature for $Ni_{1-x}P_x$.

FIG. 6. Log-log plot of the thermoelectric power vs temperature for the $x=0.202$ and 0.243 samples. The straight lines $\ln Q = \ln C + B \ln T$ are fitted to our data above 80 K [$C(x=0.202)=-3.262$, $B(x=0.202)=0.704$, $C(x=0.243)=-2.932$, and $B(x=0.243)=0.763$].

FIG. 7. Average second derivative of $Q(T)$ $\langle d^2Q/dT^2 \rangle$ in picovolts per cubic degrees Kelvin vs phosphorus atomic fraction.

from positive to negative, and the thermoelectric power increases smoothly from negative to positive. The decrease of α with ρ follows the Mooij correlation; α changes sign near $\rho = 150$ $\mu\Omega$ cm, the same resistivity value which appears in that correlation. The change in sign of the temperature coefficient of resistivity as the composition is varied has already been observed in Ni$_{1-x}$P$_x$ by Cote[9] as well as in many other metallic glass systems.[12]

What makes our results on the Ni$_{1-x}$P$_x$ system special is that the phosphorus content can be varied sufficiently so that as x is changed, the behavior of Q as well as of ρ and α smoothly spans the variety of electrical transport phenomena seen in many different metallic glasses. The resistivity of Ni$_{1-x}$P$_x$ near $x=0.15$ is low (100 $\mu\Omega$ cm), α is positive, and Q is negative as is also seen in the metallic glass Pd$_{0.775}$Cu$_{0.06}$Si$_{0.165}$. The resistivity of Ni$_{1-x}$P$_x$ with $x=0.243$ is high (170 $\mu\Omega$ cm), α is negative, and Q is positive as is the case in many high-resistivity metallic glasses [e.g., Be$_{0.4}$Ti$_{0.5}$Zr$_{0.1}$, Cu$_{0.5}$Zr$_{0.5}$, or (Ni$_x$Pt$_{1-x}$)$_{0.75}$P$_{0.25}$ with $x \leq 0.50$]. Positive thermoelectric power is a general feature of metallic glasses with high resistivity and negative Q is characteristic of low-resistivity metallic glasses. In Ni$_{1-x}$P$_x$ we see the thermoelectric power increase monotonically with the resistivity: At the lowest resistivities $Q<0$ and at higher resistivities $Q>0$. Just as Ni$_{1-x}$P$_x$ is an example of the Mooij correlation between α and ρ, this alloy system also exhibits a correlation between Q and the size of ρ.

The dependence on composition of the resistivity, the temperature dependence of the resistivity, and the size of the thermoelectric power can be interpreted in terms of the Ziman theory of electron scattering. As the phosphorus content increases the average valence of the alloy also increases so that
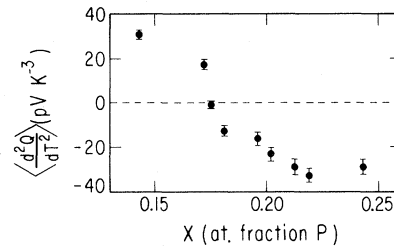
$2k_F$ will increase with increasing x. When $2k_F$ is approximately equal to k_p, the resistivity will be large [since $\rho \propto S(2k_F)$], α will be negative, and Q will be positive. Thus for the sample $x=0.243$, we find the largest values of ρ and Q and the most negative value of α for all the samples studied. For the lowest values of x, $2k_F < k_p$ and ρ will be small, α will be positive, and Q negative. This is seen in the alloys $x=0.143$ and 0.172. Thus as phosphorus content increases, ρ increases, α decreases and changes from positive to negative, and Q increases from negative to positive.

Note that the compositions where $\alpha=0$ and $Q=0$ are not the same because of the differing dependences of $\rho(T)$ and Q on the position of $2k_F$ relative to k_p. From Eq. (1) it can be seen that $\alpha=0$ when $S(2k_F)=1$, and from Eq. (5) $Q=0$ when $q = \frac{3}{2} - \frac{1}{4}r$.

Our results contradict the prediction of Cote and Meisel for the compositional dependence of the thermoelectric power. Using Ziman theory and an estimate of the contribution to the effective valence of 0.5 electrons per Ni atom and 5 electrons per P atom, they calculated the same compositional dependence of ρ and α as we see, but predicted a slow decrease in the magnitude of Q (300 K) from 3.20 μV/K at $x=0.15$ to 2.51 μV/K at $x=0.25$.[40] A somewhat lower estimate for the phosphorus contribution to the effective valence will place the Fermi wave vector lower with respect to the first peak in the structure factor and the Ziman theory would then predict an increasing thermoelectric power as a function of x as we see in our data. Given the lower effective valence, however, Cote and Meisel's estimate for the resistivity and its temperature coefficient would not agree as well with the measured values. In Ni$_{0.81}$P$_{0.19}$, Esposito, Ehrenreich, and Gelatt[41] calculate an even larger value for the effective valence $z^* \approx 2$ corresponding to $2k_F > k_p$. Their Ziman-theory calculation of the resistivity gives a value in fair agreement with experiment, 169 $\mu\Omega$ cm, but implies a negative thermoelectric power

on the order of $-5\ \mu V/K$.

As Espositio *et al.* have emphasized and as comparisons between calculated and experimental values show, the transport properties depend sensitively on the free parameters in the Ziman theory, especially E_F and $2k_F$. The Ziman theory gives the qualitative dependence of ρ, α, and Q on composition in $Ni_{1-x}P_x$ with many other metallic glass systems. The calculated magnitudes of the transport properties are only roughly correct, however. In addition, the Ziman theory is a weak scattering perturbative theory. Thus its applicability is questionable in metals in which the electron mean free path is very short.

The Ziman theory is consistent with the Mooij correlation in predicting a decrease in α as ρ increases in any particular alloy system. However, it does not predict the universal dependence (from one alloy system to another) of the sign and magnitude of α on the magnitude of ρ. In particular, it does not indicate why the value $\rho=150\ \mu\Omega$ cm should be the crossover between positive and negative values of α.

Results similar to the Ziman theory have been obtained without assuming weak scattering or needing to define $2k_F$. Nicholson and Schwartz nonperturbatively calculated the effect on the electronic properties in a disordered metal by the first peak in the structure factor.[42] They found a minimum in the electronic density of states which was correlated to the position of the first peak in the structure factor. The minimum in the density of states produces a maximum in the resistivity as a function of energy. Raising the temperature will decrease the peak of the structure factor and the minimum in $N(E)$ will not be as deep. Systems with E_F near the minimum in $N(E)$, i.e., those with high resistivities, should have negative temperature coefficients of resistivity. These results are very similar to what happens in the Ziman theory when $2k_F$ is near k_p. This calculation indicates, therefore, that the qualitative features predicted by the Ziman theory may actually be relevant to metallic glasses in the highly resistive regime. This theory, along with the localization models of Jonson and Girvin and Imry are the only ones explicitly suited to strong scattering systems.

On the negative side, the Ziman theory predicts a linear dependence of the thermoelectric power on T, and as we have seen none of the thermoelectric powers we measured are linear at all temperatures (Fig. 5). At the lowest phosphorus concentrations ($x=0.143$, 0.172, and 0.175) the thermoelectric powers are not even a monotonic function of T and are reminiscent of those seen in iron-based ferromagnetic metallic glasses.[2] In the samples of higher P concentration ($x>0.18$), $Q(T)$ is monoton-

ic and has less structure in its temperature dependence.

The positive thermoelectric power of high-resistivity metallic glasses such as $Be_{0.4}Ti_{0.5}Zr_{0.1}$ ($\rho=300\ \mu\Omega$ cm) or $Cu_{0.5}Zr_{0.5}$ ($\rho=200\ \mu\Omega$ cm) are very linear over a very wide range of temperatures. The negative thermoelectric power of $Pd_{0.775}Cu_{0.06}Si_{0.165}$ ($\rho=85\ \mu\Omega$ cm) departs markedly from linearity. The straying from the linearity in nonmagnetic metallic glasses with low resistivity may be explained by hypothesizing another scattering mechanism, and using the Nordheim-Gorter relation. Thus in high-resistivity metals where the scattering is dominated by the disorder, the thermoelectric power is quite linear. In low-resistivity metals the contribution from disorder to the resistivity is less and other scattering mechanisms such as s-d scattering can make a larger proportional contribution to $Q(T)$.

The observed transport behavior cannot be explained by the Mott s-d scattering model over the entire composition range. Low-temperature heat-capacity measurements in $Ni_{1-x}P_x$ indicate that as x increases from 0.15 to 0.25, $N(E_F)$ decreases by a factor of 3.[43] The s-d model predicts that ρ will decrease as x increases since $\rho \propto N_d(E_F)$ and that Q will be negative since $Q \propto [d(\ln N_d)/dE]_{E_F}$. The measured resistivity increases with x, however, and Q changes sign in the middle of the composition range.

It has been suggested that Mott s-d scattering may contribute to the resistivity at low-phosphorus concentrations where the d band is not full.[44] The resistivity in this composition range is fairly low, and, therefore, s and d electrons may have different mobilities. Mott s-d scattering in a metal with a nearly full d band produces a negative contribution to the thermoelectric power. The negative thermoelectric power we find below $x=0.18$ is consistent with the existence of Mott s-d scattering. The Nordheim-Gorter relation states that scattering mechanisms contribute to the thermoelectric power in proportion to their contribution to the resistivity.[26] Thus in our low-phosphorus alloys in which the resistivities are low the negative thermoelectric power produced by Mott s-d scattering is likely to be significant. As x is increased, $N_d(E_F)$ decreases, and so the s-d scattering contribution to the resistivity and negative contribution to the thermoelectric power will decrease. Therefore, the thermoelectric power should become more positive as x is increased which is what we observe. The decrease in Mott s-d scattering in competition with an increase in disorder scattering with increasing x could explain insensitivity of the resistivity to changes in x in the range $x<0.18$ (see Fig. 2).

The two-level system scattering model of Tsuei can account for a negative α in $Ni_{1-x}P_x$ with $x=0.243$. His structural model for the two-level systems in transition-metal—metalloid metallic glasses predicts an increase in the number of such systems as the metalloid concentration increases above 20%.[22] The contribution to the resistivity from electron scattering by two-level systems increases as the number of two-level systems increases. Therefore, with increasing metalloid content ρ will increase, and, since electron-two-level system scattering decreases with temperature, α will decrease. A major objection to applying the two-level system model to explain the existence of a negative α at high temperatures in metallic glasses has been the absence of corresponding anomalies in $Q(T)$ at those temperatures.[1] Instead, $Q(T)$ is very linear at high temperatures in metallic glasses with negative α such as $Cu_{0.5}Zr_{0.5}$. In $Ni_{0.757}P_{0.243}$, $Q(T)$ is certainly nonlinear but it is difficult to see why two-level system scattering would cause anomalies in $Q(T)$ for this glass but not in $Cu_{0.5}Zr_{0.5}$ if they are causing a negative α in both glasses.

Since the dependence of α on ρ follows the Mooij correlation, our resistivity data are consistent with the localization theories of Jonson and Girvin[23] and of Imry.[24] These theories give no explanation for the compositional dependence of ρ or Q, however, nor do they explain the correlation of positive Q with large values of ρ.

Several explanations have been proposed for small deviations from perfect linearity in $Q(T)$. The thermoelectric power of many metallic glasses is linear over a wide range of temperatures but extrapolates to a positive value at $T=0$. Closer examination at low temperatures by Gallagher revealed that the slope of $Q(T)$ was greater than at high temperatures.[45] He attributed the enhanced slope at low temperatures to electron-phonon mass enhancement, significant when $T \ll \Theta_D$. Not all nonmagnetic metallic glasses have enhanced slopes at low temperature, however. $Ni_{0.36}Zr_{0.64}$ and $Ca_{0.8}Al_{0.2}$ (Ref. 8) have linear $Q(T)$ at a high temperature which extrapolate to negative values of $Q(0)$, and $Cu_{0.5}Zr_{0.4}Fe_{0.1}$ has a linear $Q(T)$ within error from well below 50 to about 250 K.[45] In the present case, the low-temperature slope is larger than at higher temperatures, but the high temperature $Q(T)$ is not linear even allowing for a positive intercept at $T=0$ (Fig. 5). Another explanation was given in the case of $Ni_{0.76}P_{0.24}$ by Cote and Meisel.[28] Their measurement of the thermoelectric power showed a lower slope above 300 K than below that temperature and their data were linear in T between 80 (the lowest temperature measured) and 300 K, extrapolating to near zero at $T=0$. They explained the difference in

slope by the Pippard-Ziman condition: Phonons with wavelengths longer than the electron mean free path are ineffective electron scatterers. Their calculation, incorporating this into the Ziman theory, predicted a decrease in the magnitude of the slope of $Q(T)$ at high temperatures. Our data show such an effect for all compositions except $x=0.175$ and 0.181; the magnitude of the slope of $Q(T)$ continuously decreases with increasing temperature. For example, in our samples with $x>0.20$ we see a decrease in dQ/dT of approximately 20% between the temperature range $100 < T < 300$ K and the range $300 < T < 400$ K. Quantitative comparison with the calculation of Cote and Meisel is difficult since the parameters of the theory are hard to determine, but a 20% decrease seems to be of the correct order of magnitude. On the other hand, metallic glasses with higher resistivities such as $Cu_{0.5}Zr_{0.5}$ or $Be_{0.4}Ti_{0.5}Zr_{0.1}$ do not show the predicted decrease in the slope of $Q(T)$ above 100 K.

The decrease of structure in $Q(T)$ as P content increases may also be related to the concurrent change of ferromagnetic properties in amorphous $Ni_{1-x}P_x$. The structure of $Q(T)$ changes around $x=0.18$, the composition below which ferromagnetism increases rapidly with decreasing x. The second derivative of $Q(T)$ decreases with increasing x and changes sign at that composition. As already mentioned, these changes may be caused by s-d scattering below $x=0.18$ where the d band is not yet full. The similarity of the temperature dependence of ρ and Q in this range of compositions with those seen in iron-based ferromagnetic metallic glasses suggests that they may have a magnetic origin. The structure in $\rho(T)$ and $Q(T)$ of iron-based ferromagnetic metallic glasses is highly sensitive to the addition of small amounts of a transition element which also changes the magnetic properties. For example, 2% Mo in $Fe_{0.80}B_{0.20}$ decreases the Curie temperature from 647 to 595 K, changes the single minimum in $\rho(T)$ at $T=14$ K to two minima at 7 and 80 K,[5] and washes out much of the structure seen in $Q(T)$ of $Fe_{0.80}B_{0.20}$.[2] Equally dramatic differences are seen between $Fe_{0.40}Ni_{0.40}P_{0.14}B_{0.06}$ ($T_C=537$ K) and $Fe_{0.32}Ni_{0.36}Cr_{0.14}P_{0.12}B_{0.06}$ ($T_C=250$ K). Without the Cr, the minimum of $\rho(T)$ is at $T=26$ K, but the minimum moves to $T=270$ K with Cr.[5] The thermoelectric power of $Fe_{0.40}Ni_{0.40}P_{0.14}B_{0.06}$ is negative below $T=450$ K and has minimum near $T=250$ K, while $Q(T)$ of $Fe_{0.32}Ni_{0.36}Cr_{0.14}P_{0.12}B_{0.06}$ is monotonic and positive above 150 K.[2]

The system $Ni_{1-x}P_x$ and these iron-based metallic glasses show a common decrease in the structure which appears in their thermoelectric powers as the magnetic properties are varied whether by changing the relative concentrations as in $Ni_{1-x}P_x$ or by the

addition of a different magnetic element as in the Fe containing metallic glasses.

The $Ni_{1-x}P_x$ system can be compared with the ferromagnetic metallic glass system $Fe_{1-x}B_x$. The Curie temperature in $Fe_{1-x}B_x$ increases slowly with increasing x from 509 K at $x=0.125$ to 647 K at $x=0.20$ (Metglas 2605).[46] The temperature coefficient of resistivity is positive and decreases as x increases from 0.13 to 0.16 and remains approximately constant in the range $0.16 < x < 0.26$.[47] The thermoelectric power is negative and approximately constant[48] above 300 K similar to the $Ni_{1-x}P_x$ samples with $x=0.143$ and 0.172. The value of Q near room temperature has a smooth minimum as a function of x near $x=0.20$. The compositional dependence of T_C, α, and Q is much weaker in $Fe_{1-x}B_x$ than in $Ni_{1-x}P_x$. The magnitude of ρ, the positive α, and the negative and nonmonotonic $Q(T)$ seen in $Fe_{1-x}B_x$ is very similar to what is seen in $Ni_{1-x}P_x$, $x=0.143$ and 0.172. Two interesting differences can be seen though. The Curie temperature decreases with increasing x in $Ni_{1-x}P_x$ but increases with x in $Fe_{1-x}B_x$. The thermoelectric power increases with x in $Ni_{1-x}P_x$ and becomes positive for $x > 0.18$ while Q decreases with x in $Fe_{1-x}B_x$ becoming more negative. As already mentioned, the features of $\rho(T)$ and $Q(T)$ are dramatically altered by 2% Mo as in $Fe_{0.78}Mo_{0.02}B_{0.20}$. The effects on the $Ni_{1-x}P_x$ system of adding such elements may also be quite interesting.

Finally, we can consider the effect of the preparation technique on the transport properties. The major difference between the quenched and chemically deposited samples was in the temperature dependence of the temperature coefficient of resistivity, $\alpha(T)$ (Fig. 4). In the chemically deposited samples, $\alpha(T)$ has a maximum near 80 K, while in the quenched samples $\alpha(T)$ increases above 80 K. The thermoelectric power, on the other hand, seems to be insensitive to the preparation technique. No corresponding differences in the temperature dependence of Q are obvious between the quenched samples with $x=0.202$ and 0.213 and the deposited samples with $x=0.196$ and 0.219 (Fig. 5). The quenched $x=0.181$ sample and the deposited $x=0.175$ sample are in the composition range where $Q(T)$ changes both its sign and its curvature as x is varied. In both samples, $Q(T)$ has negative curvature at low temperature and positive curvature above about 200 K.

The difference in $\alpha(T)$ could indicate differences in the local atomic order between quenched and deposited glasses. Using ^{31}P NMR measurements of the Knight shift K and the field-dependent

linewidth parameter k_1, Bakonyi et al. found greater inhomogeneities among ^{31}P sites in $Ni_{1-x}P_x$ glasses prepared by chemical deposition from a chloridic bath than in those prepared by dc electrodeposition and melt quenching.[34] Lashmore et al.[49] found two distinct dependences of K on x in $Ni_{1-x}P_x$, $0.10 < x < 0.25$. Their results in samples prepared by chemical deposition, melt quenching, and pulse electrodeposition are very similar to those seen by Bakonyi et al. in the same composition range, K decreases with increasing x, but in their dc electrodeposited samples K is lower and changes little with x in the same range. Lashmore et al. attribute the difference in $K(x)$ to two distinct amorphous structures and note that by annealing the samples prepared by the other methods K changes to a value similar to that seen in the dc electrodeposited samples of the same composition. The existence of two amorphous structures is also supported by the difference in crystallization temperature between Cote's electrodeposited samples and our melt quenched and chemically deposited samples. Cote found that the crystallization temperature decreased monotonically with increasing x from 657 K at $x=0.154$ to 557 K at $x=0.254$.[37] We found a maximum in the crystallization temperature of 630 K at $x=0.202$.

In conclusion, we have measured the temperature dependence of ρ and Q in a single metallic glass system $Ni_{1-x}P_x$. We find that as x increases ρ increases, α decreases and becomes negative, and Q increases and becomes positive. These results are in agreement with correlations between ρ and α and between ρ and Q seen in many metallic glasses. At low-phosphorus concentrations, $x < 0.18$, the transport properties are similar to those seen in iron-based metallic glasses, and as x increases above 0.18, the transport behavior becomes more similar to what is seen in nonmagnetic metallic glasses. The dependence on x of ρ, α, and the size of Q are qualitatively consistent with the Ziman theory although it seems probable that other scattering mechanisms, such as s-d scattering, are important in the samples with low P concentration. There is a clear effect of the preparation technique on the transport properties only in the temperature dependence of α.

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