

## Temperature dependence of the resistivity of Nb:Ni glasses\*

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(Received 26 July 1977)

The temperature dependence of the resistivity  $\rho$  of  $\text{Nb}_{1-c}\text{Ni}_c$  metallic glasses is reported as a function of concentration in the range  $c = 0.4$  to  $c = 0.6$ . This spans the interval in which glasses can be quenched from the melt in this system. We find  $\alpha \equiv (1/\rho)(d\rho/dT) < 0$  for all samples studied. We interpret these results in terms of a recent calculation which extends the Ziman theory to metallic glasses. In order to fit the data with this theory one must assume that  $2k_F \simeq k_p$ , where  $k_p$  is the position of the first peak in the structure factor. Good agreement between the theory and the experiment is found if electron-electron scattering is also included. The fact that  $\alpha < 0$  for all glass forming compositions in this system tentatively supports an extension of an electronic model of glass formation proposed for the transition-metal-metalloid systems to these amorphous intertransition-metal alloys.

### I. INTRODUCTION

Although there has recently been considerable interest in metallic glass alloys, the nature of the transport properties, and of the glass formation itself, has remained controversial. The availability of various types of glasses enables one to establish which properties depend to a large extent only on the fact that the metal is amorphous and which are specific to a particular alloy composition. Most work until recently has concentrated on one type of glass which has the form  $M_c X_{1-c}$  where  $M$  is a noble or transition metal with a nearly filled  $d$  band and  $X$  is an element of group IVA or VA. In these alloys,<sup>1</sup> glass formation occurs only in a restricted range of composition with the metalloid content near 20 at.%, as in  $\text{Pd}_{0.8}\text{Si}_{0.2}$  for example. Photoemission measurements<sup>2</sup> indicate that these glasses have a minimum in the density of states,  $D(E)$ , at the Fermi level. Resistivity studies<sup>3</sup> show that these glasses have a small and often negative value of the temperature coefficient of resistivity,  $\alpha \equiv (1/\rho)(d\rho/dT)$ , whereas for most crystalline metals  $\alpha$  is large and positive. It is of interest to see if these properties are also found in other types of glasses such as the intertransition-metal alloys,  $N_{1-c}M_c$ , where  $N$  is a transition metal with a  $d$  band less than half full and  $M$  is as defined above. These glasses generally crystallize<sup>4</sup> at much higher temperatures than do those containing metalloids. Another important difference is that these alloys typically form glasses over a wide range of composition centered about 50%. The transport properties have until now not been studied as a function of composition.

A recent photoemission<sup>5</sup> study indicated that for glasses from one such system, Nb:Ni, the density

of states at the Fermi level,  $D(E_F)$ , was near a minimum for several glass-forming compositions. In the present paper we report a study of the temperature dependence of the resistivity for several Nb:Ni glasses that span the glass forming composition range. For all the compositions studied it was found that  $\alpha < 0$ . These results, when combined with the photoemission data, suggest that there may be a common mechanism underlying the glass formation in both the intertransition-metal and transition-metal-metalloid alloys. This model of glass formation<sup>6,7</sup> in which the electronic states are treated by perturbation theory, was originally proposed only for the latter type of glass and relates the minimum at  $D(E_F)$  to the negative value of  $\alpha$ . The temperature dependence of the data is also considered in terms of a recent theory<sup>8</sup> of resistivity for disordered metals. This theory extends the Ziman theory for liquid metals into the solid state. However, in order to fit the present data by this theory an additional term must be included which has a possible explanation in terms of the  $d$  bands at the Fermi surface. Data taken on a Nb:Ni alloy in the liquid state<sup>3</sup> have a similar temperature dependence as our data and support the idea that the theories which have been developed for describing the liquid state can be extended to treat these metallic glasses.

In Sec. II, we will present the experimental results and in Sec. III discuss these results in terms of the theories mentioned above.

### II. RESULTS

Alloys of Nb and Ni can be quenched<sup>4</sup> into the glass state in the region between 40 and 66-at.% Ni. The samples used in this experiment,

$\text{Nb}_{0.4}\text{Ni}_{0.6}$ ,  $\text{Nb}_{0.5}\text{Ni}_{0.5}$  and  $\text{Nb}_{0.6}\text{Ni}_{0.4}$ , span almost this entire range. As described elsewhere,<sup>4</sup> the samples were prepared in an identical manner as the samples used in the photoemission study<sup>5</sup> by rapid arc furnace quenching<sup>9</sup> of the melt. The samples were approximately 30- $\mu\text{m}$  thick and were cut into thin strips 1.5-cm long and 0.1-cm wide. The resistivity was measured using a four-probe ac null technique with a driving frequency of 95 Hz in order to eliminate the effects of voltages due to thermal gradients. Contacts to the sample were made with silver epoxy and were heated to 200°C for 1 h in order to ensure good contact. This temperature is well below the crystallization or annealing temperature of the glass. The samples were then mounted in a helium Dewar where the temperature could be varied from 4.2 to 360 K. Although the absolute magnitude of the resistivity is difficult to determine due to errors in accurately measuring the size of the samples, the temperature coefficient of resistivity  $\alpha$  can be determined much more precisely because the geometrical factors cancel out. We estimate our error in measuring the absolute resistivity as  $\pm 15\%$  and in measuring resistivity ratios as  $\pm 0.02\%$ .

For all three samples the magnitude of the resistivity is the same within our estimated uncertainty and was  $\rho = 150 \pm 22 \mu\Omega \text{ cm}$ . In Fig. 1 we show the temperature variation of the resistivity for the three samples. Each curve is normalized to the value of the resistivity at 300 K. As can be

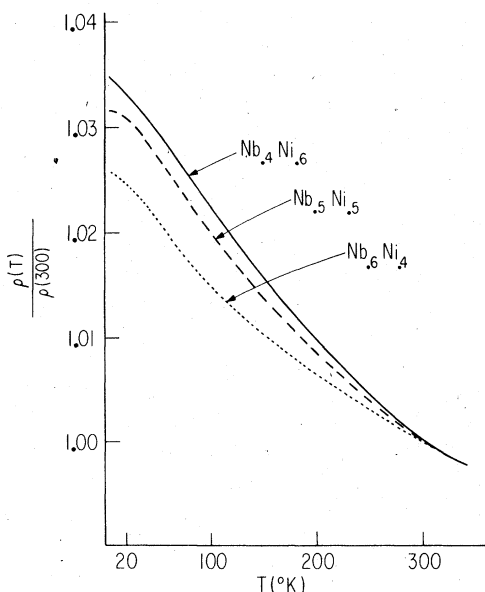


FIG. 1. Resistivity, normalized to the room-temperature value, as a function of temperature for the three glass samples.

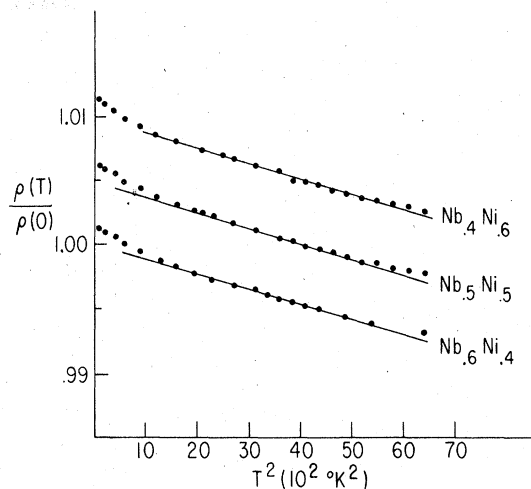


FIG. 2. Low-temperature resistivity, normalized to an extrapolated zero-temperature value plotted vs  $T^2$ . The three curves are displaced from each other vertically by 0.005. Note that the slopes of the three curves are the same.

readily seen, this variation is anomalous for a metal since the resistivity decreases as the temperature is increased. Also the change of the resistivity with temperature is quite small, varying by only  $\sim 3\%$  between helium and room temperature. The low-temperature behavior is shown more clearly in Fig. 2. In this figure, the normalized resistivity is plotted vs  $T^2$ . The slopes of the curves are virtually identical for each of the samples and fit a quadratic function in the region between 25 and 80 K. In this region the data can be fit by the equation  $\rho(T) = \rho_0 [(1 - 1.23) \times 10^{-6} T^2]$ , where  $T$  is the temperature in degrees Kelvin. Both below 25 K and above 80 K the experimental points lie higher than is predicted by this relation. We will discuss the possible origins of this behavior in the next section. However it is clear that since the low-temperature behavior of the three samples is the same, in order to account for the differences seen in Fig. 1, one must concentrate on the behavior of the resistivity at temperatures greater than 80 K. This suggests that there may be a variety of physical mechanisms which have a role in determining the resistivity and that these can be distinguished by their relative importance in different temperature regimes.

### III. DISCUSSION

There has been considerable controversy about how to understand the resistivity data on glasses. One model<sup>10, 11</sup> that has been proposed is based on the tunneling of atoms in a disordered solid between two sides of a double potential well. This model predicts that the resistivity should behave

in the same manner as would be predicted by the Kondo mechanism of scattering from magnetic impurities<sup>12</sup> and would have a region where the resistivity varies as  $\ln T$

$$\rho(T) = A + B\rho_N(T) - C \ln(T^2 + D), \quad (1)$$

where  $A$ ,  $B$ ,  $C$ , and  $D$  are constants and  $\rho_N(T)$  is the regular electron-phonon term. Unfortunately it is very difficult to determine *a priori* the parameters of the model including the parameter  $C$ , which is a measure of how many tunneling states exist in a glass. It is therefore impossible to predict *a priori* the size and the temperature dependence of the electrical resistivity. In many alloys where this model has been used to interpret the resistivity data,<sup>11</sup> it was assumed that the number of tunneling states depended strongly on the relative concentration of the various elements [e.g.,  $(\text{Ni}_{0.5}\text{Pd}_{0.5})_{1-x}\text{P}_x$ ]. For the alloys we have studied, Nb:Ni, the low-temperature behavior is essentially independent of concentration. Since there is a considerable atomic size difference between Nb and Ni, we would expect a large change in the number of tunneling states when the concentration is changed by 20%. We conclude, therefore, that this mechanism is not primarily responsible for the anomalous negative values of  $\alpha$  found in these glasses. Furthermore, the absence of a concentration dependence to  $\alpha$  implies that it is unlikely that the negative slope arises from a magnetic effect involving the Ni spins such as the Kondo effect<sup>12</sup> or scattering from spin clusters.<sup>13</sup>

An alternative to assuming the existence of tunneling states in these glasses, is to focus attention on the normal electron-phonon contribution to the resistivity,  $\rho_N(T)$ . Clearly this contribution will be strongly modified by the absence of periodic order. We show here that an extension of the Ziman theory,<sup>14</sup> which was originally developed to explain the resistivity behavior of simple liquid metals, can tentatively be used for these alloys. One of the early achievements of this theory was that it could explain the negative temperature coefficients in the divalent metals. The formalism was subsequently extended by Evans *et al.*,<sup>15</sup> to treat the case of liquid transition metals. This has given good agreement with the experimental values of the resistivity for many transition metals.<sup>16</sup> That this theory could be applied to metallic glass alloys was originally suggested by Sinha.<sup>17</sup>

In a recent paper<sup>8</sup> one of us has calculated what the temperature dependence of the resistivity should be for an amorphous solid in terms of this theory. The final expression for the resistivity is

$$\rho(T) = \frac{30\pi^3\hbar^3}{me^2k_F^2E_F\Omega} \sin^2[\eta_2(E_F)] \times \{1 + [S_0(2k_F) - 1] e^{-2[W(t) - W(0)]}\}, \quad (2)$$

where  $k_F$  and  $E_F$  are the Fermi wave vector and energy, respectively,  $\Omega$  is the atomic volume.  $\eta_2(E_F)$  is the  $d$ -wave phase shift describing the scattering of the conduction electrons, of energy  $E_F$ , by the ion cores which each carry a muffin-tin potential. The temperature dependence enters through the Debye-Waller factor  $W(T)$ . In the Debye approximation we have

$$W(T) = \frac{3\hbar^2k^2T^2}{2Mk_B\Theta^3} \int_0^{\Theta/T} \left( \frac{1}{e^z - 1} + \frac{1}{2} \right) z dz, \quad (3)$$

where  $\Theta$  is the Debye temperature and we will evaluate the expression at  $k = 2k_F$ . For  $T \geq \Theta$ ,  $\rho(T)$  varies as a linear function of  $T$  and for  $T \ll \Theta$  it varies quadratically with  $T$ . Equation (2) can easily be modified for the case of a binary alloy.<sup>14</sup>

As we have already mentioned the experimental data, as seen in Fig. 2, varies quadratically with  $T$  at low temperatures. This is in agreement with the predictions of this theory. In terms of this theory the fact that the resistivity decreases with increasing temperature implies  $S(2k_F) > 1$  or that  $2k_F$  is near the first peak in the structure factor. Assuming,<sup>8</sup> therefore, that  $S(2k_F) \sim 3$ ,  $2k_F \sim 3 \text{ \AA}^{-1}$ , and the mass is the average of the two elements, we find that in order to fit the experimental data in this low-temperature region we must use  $\Theta = 248 \pm 5 \text{ K}$ . These parameters are as stated above for all three samples since in the low-temperature region the slopes are the same. The value of the Debye temperature is quite a reasonable one: it is within 10% of the value<sup>18</sup> for pure Nb and 32% less than the average of the Debye temperatures of the component elements. For comparison, the Debye temperature found<sup>19</sup> from specific-heat data on a similar binary intertransition-metal glass,  $\text{Zr}_{0.43}\text{Cu}_{0.57}$ , is 200 K which is 32% lower than the average of the Debye temperatures of the two component elements.

Although the theoretical predictions are in good agreement with the experimental data at low temperatures, at temperatures higher than 100 K there is considerable discrepancy. In Fig. 3 we show the data for the  $\text{Nb}_{0.4}\text{Ni}_{0.6}$  sample (dashed line) as well as the theoretical curve (solid line) derived from Eq. (2) with the parameters chosen as stated above. At low temperatures, the agreement is good since the parameters other than the exponent of  $T$  were chosen to fit this region. At higher temperatures the experimental curve decreases less rapidly than is predicted. There is

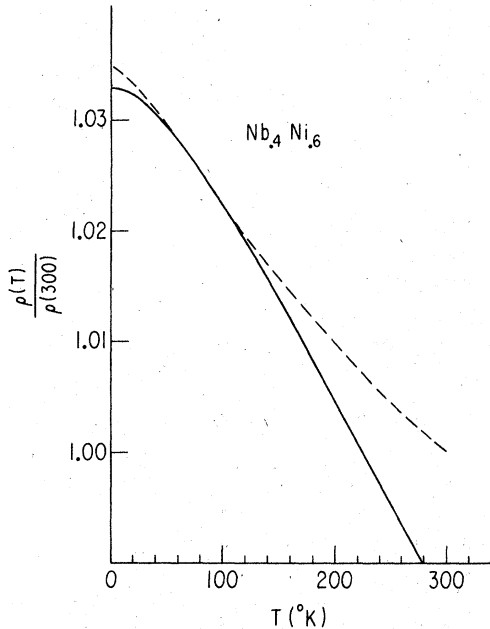


FIG. 3. Measured resistivity of  $\text{Nb}_{0.4}\text{Ni}_{0.6}$  (dashed curve) compared with the theoretical resistivity (solid curve) determined from Eq. (2) with the parameters mentioned in the text. Note the deviation between experiment and theory at high temperatures.

possibly another scattering mechanism which is important in these intertransition-metal alloys which does not contribute<sup>8</sup> in the transition-metal-metalloid glasses such as  $(\text{Pd}_x\text{Cu}_{1-x})_{0.8}\text{P}_{0.2}$ . If we subtract the two curves in Fig. 3 we find that this contribution varies as  $T^2$  as is shown in Fig. 4. We see that for all three samples this additional contribution varies roughly quadratically but has a different slope for each sample.

One hypothesis for the origin of this term that is

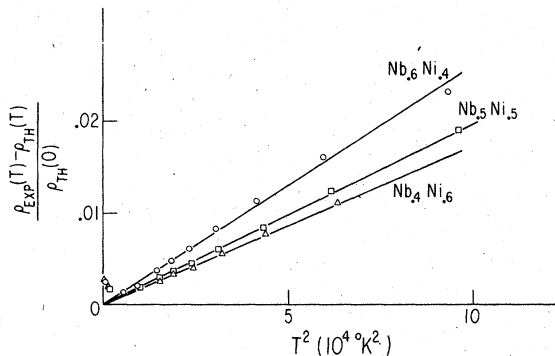


FIG. 4. Deviation between the two curves plotted in Fig. 3 vs  $T^2$ . Similar curves are plotted for the other two samples studied. Note that the deviations of the experimental curve from the theoretical one is quadratic in  $T$  for all three samples.

consistent with all the data is electron-electron scattering between the  $s$  and  $d$  electrons. It is well known<sup>20</sup> that electron-electron scattering will increase quadratically with temperature. The contribution of electron-electron scattering in these alloys should be comparable with that found in a pure transition metal. In pure palladium, it was found<sup>21</sup> that the contribution of  $e-e$  scattering at room temperature was  $\sim 2.7 \mu\Omega \text{ cm}$ . For our samples we find at 300 K contributions of  $2.5 \mu\Omega \text{ cm}$  ( $\text{Nb}_{0.4}\text{Ni}_{0.6}$ );  $2.8 \mu\Omega \text{ cm}$  ( $\text{Nb}_{0.5}\text{Ni}_{0.5}$ ); and  $3.8 \mu\Omega \text{ cm}$  ( $\text{Nb}_{0.6}\text{Ni}_{0.4}$ ) which are all comparable to that value. Furthermore in the intertransition-metal alloys we expect that the Fermi level is in the middle of the  $d$  bands<sup>5</sup> so that  $s-d$  electron-electron scattering can occur. However, for those glasses containing metalloids, such as  $\text{Pd}_{0.8}\text{Si}_{0.2}$ , the Fermi level lies above the  $d$  bands where the electronic states are of  $s$  or  $p$  character.<sup>2,22</sup> This can explain why there is a contribution from this effect in the samples used in the present study but not in those containing metalloids, where Eq. (2) seems to adequately fit the data over the entire range of temperature measured.<sup>8</sup>

One final discrepancy between the data and the theory is at very low temperatures  $T < 20 \text{ K}$ . In this region the resistivity increases rapidly as the temperature is lowered. We believe that this is due simply to the presence of magnetic impurities which give rise to a Kondo term in the resistivity.<sup>12</sup>

We have been able to satisfactorily explain the resistivity data on these glasses in terms of an extended Ziman theory if electron-electron scattering processes are included. In the derivation<sup>8</sup> of Eq. (2) a term was dropped which was argued to be unimportant [the last term in Eq. (8) of Ref. 8]. In the present study, good agreement with experiment was obtained without the inclusion of the term. It should also be mentioned that the resistivity data on a liquid Nb:Ni sample has a similar temperature dependence as in the glass.<sup>3</sup> This result strengthens the application of the Ziman theory, originally developed for explaining the resistivity behavior in liquids, to include glasses as well. The results of an independent study<sup>23</sup> of the resistivities of Nb:Ni and other glasses above 300 K and the corresponding liquids will be reported elsewhere.

A central assumption that has enabled us to use this theory of resistivity was that  $2k_F$  was near  $k_p$ , a peak in  $S(k)$ , in these alloys. If this is indeed true one would expect also to see a minimum in the density of states at the Fermi energy.<sup>6</sup> In fact evidence for this can be found by observing the photoemission spectra of Nb:Ni glasses<sup>5</sup> where a dramatic decrease is found in the density of states

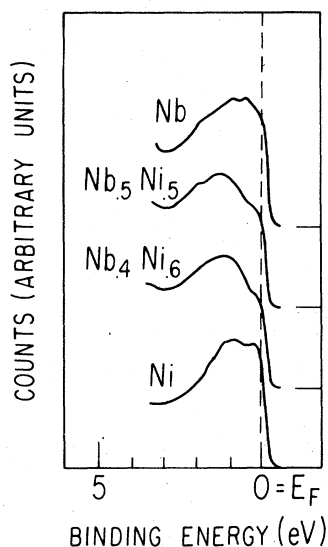


FIG. 5. Photoemission spectra, taken from Ref. 5, for two of the glasses studied as well as for pure Ni and pure Nb. Note that in both glasses there is a drop in  $D(E)$  at the Fermi level whereas for both pure elements the density of states at  $E_F$  is close to its maximum value.

in the region around the Fermi level. The low-binding-energy spectra are shown in Fig. 5 for two of the samples as well as for pure Nb and pure Ni. Whereas for the pure elements  $D(E)$  is near a maximum at  $E_F$ , for both glasses the maximum in  $D(E)$  has shifted to  $\sim 1.5$  eV below  $E_F$ . If the resistivity measurements are also considered, the negative temperature coefficient of resistivity indicates that  $2k_F \sim k_p$  for the entire glass-forming region.

The remaining difficulty is to understand why  $2k_F \sim k_p$  in these alloys. There have been no band-structure calculations for alloys of this type so that it is unclear how the rather narrow, nearly filled  $d$  band of Ni will interact with the wider and less than half-filled  $d$  band of Nb. Whereas in alloys containing metalloids an effective valence could be assumed, there is less evidence, if any, that a similar procedure can be used in the case of an intertransition-metal alloy such as Nb:Ni. However calculations<sup>24</sup> of the wave functions for the lowest-energy  $d$  bands of a transition metal show that these states are diffuse and do not resemble atomic  $d$ -wave-functions, whereas the higher-energy bands are more like their atomic counterparts. It is perhaps plausible that elements such as Nb, with less than half of their  $d$  bands occupied, will play the same role as the metalloids upon alloying with a transition metal such as Ni, with a nearly full  $d$  band. Following Szofran *et al.*,<sup>25</sup> we may tentatively assume that each Nb

atom contributes most of its five valence electrons to the free-electron gas and that Ni contributes, as in the transition-metal-metalloid alloys, less than one electron per atom. As in the case<sup>25</sup> of Zr:Cu this gives qualitative agreement between  $2k_F$  and  $k_p$ . Also positron annihilation measurements<sup>19</sup> on Zr:Cu are consistent with this value of  $2k_F$ .

One additional point should be made which concerns the structure factor of glassy (or liquid) alloys. In these alloys there are three partial structure factors:  $S_{MM}(q)$ ,  $S_{NN}(q)$ ,  $S_{MN}(q)$ . These will each contribute substantially to the scattering of electrons: the concentration of both elements is approximately the same and both elements are transition metals with large scattering amplitudes at  $2k_F$ . This is different from the case of transition-metal-metalloid alloys.  $M_c X_{1-c}$ , where the concentration of metalloid,  $1-c$ , is small, approximately 0.2. The importance of the partial structure factor  $S_{XX}(q)$  is therefore reduced by  $(1-c)^2 \cong 0.04$  in the scattering of electrons.<sup>26</sup> Also the metalloid pseudopotential evaluated at  $2k_F$ ,  $v_X(2k_F)$ , is extremely small,<sup>27</sup> so that primarily only  $S_{MM}(q)$  is important for determining the resistivity and gaps in the density of states. By contrast, when considering the intertransition-metal alloys, all three partial structure factors are important instead of only one. Due to the size difference of the atoms each partial structure factor will have its peak in a different place while the relative weighting varies with composition. For example in  $Zr_{0.43}Cu_{0.57}$  the three peaks have been reported at<sup>28</sup>  $2.32 \text{ \AA}^{-1}$ ,  $2.76 \text{ \AA}^{-1}$  and  $3.00 \text{ \AA}^{-1}$ . Therefore if  $2k_F$  lies in the broad range spanned by these three values then the conditions for negative  $\alpha$  and a minimum in  $D(E)$  at  $E_F$  may occur. This may explain why the intertransition-metal alloys can form glasses over a wide range of composition whereas the alloys containing metalloids are restricted in their glass forming ability to a narrow range.

Finally we consider the possible implication of these findings for the glass formation itself. It is possible as already mentioned by Szofran *et al.*,<sup>24</sup> that a theory<sup>6,7</sup> of glass formation proposed for the transition-metal-metalloid glasses may also hold for these intertransition-metal alloys. In this theory it was proposed that glass formation would be more likely to occur when  $2k_F \sim k_p$ , so that there is a minimum in  $D(E)$  at  $E_F$ . In the present samples we observe both an anomalous temperature dependence of the resistivity and a minimum in the density of states at the Fermi level. If these two phenomena can indeed be explained by the same theory, it lends much more credibility to the use of this theory to explain the

glass formation itself in these intertransition-metal systems. For instance, the structure factor considerations mentioned above may explain why the intertransition-metal alloys can form glasses over a wide range of composition whereas the alloys containing metalloids are restricted in their glass forming ability to a narrow range. However, only relatively few intertransition-metal systems readily form glasses<sup>29</sup> so that a more sophisticated application of the electronic criteria presented here with consideration of other factors effecting glass formation<sup>30</sup> may be required.

In conclusion we have measured the temperature dependence of the resistivity of Nb:Ni glass over the entire glass forming region. We are able to analyze this data using an extension of the Ziman theory for liquid metals and agreement between experiment and theory is good if account is taken

of electron-electron scattering. This agreement also suggests that an electronic mechanism, previously suggested only for alloys containing metalloids, may be relevant to glass formation in these alloys as well. We have also suggested one explanation of why the glass forming region is so much broader for the intertransition-metal alloys than in those transition-metal-metalloid alloys. Finally the assumption that  $2k_F \sim k_p$ , which was necessary for explaining this data needs additional support. A band-structure calculation on a related crystal would be useful in order to verify or disprove this assumption.

#### ACKNOWLEDGMENTS

We would like to thank A. Bansil, H.-J. Güntherodt, E. Hauser, and J. Tauc for stimulating conversations.

\*Supported by NSF-MRL program at the University of Chicago and by ONR Contract No. N14-76-C-820 at Northeastern University.

†Supported in part by the Alfred P. Sloan Research Foundation.

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