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The Hall effect and the electrical resistivity in amorphous Ni–B–Si alloys

J Ivkov†, E Babić† and H H Liebermann‡

† Institute of Physics, University of Zagreb, PO Box 304, Zagreb, Yugoslavia

‡ Metglas Products, Allied-Signal Inc., Parsippany, New Jersey 07054, USA

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Abstract. The values for the normal Hall coefficient, R_0 , electrical resistivity, ρ , and the room-temperature coefficient of resistivity, α , for ten amorphous Ni–B–Si alloys, with metalloid concentration ranging from 20 to 39 at.%, are reported. R_0 and α continuously decrease with increasing metalloid content, or for the fixed metalloid content, with increasing silicon concentration; and α changes sign for $\rho \geq 140 \mu\Omega \text{ cm}$. The value of the Fermi wavevector deduced from R_0 (within the framework of the free-electron model) and its dependence on the composition of the alloy indicate that the transport properties of our alloys can be interpreted in terms of the Ziman–Faber theory. An alternative description of the variation of R_0 is also discussed.

It is well known that in Ni–metalloid metallic glasses the resistivity ρ increases while the room-temperature coefficient of the resistivity $\alpha = (1/\rho)d\rho/dT$ decreases with increasing metalloid content. For resistivities close to $150 \mu\Omega \text{ cm}$, α , as one would expect from the correlation obtained by Mooij (1973), changes sign and becomes negative. In the case of Ni–metalloid amorphous alloys (Ni–P in particular) it is argued (Cote and Meisel 1981, Carini *et al* 1983) that such behaviour, as well as electronic transport in general, can be explained in terms of the diffraction model (i.e. Ziman–Faber theory). As the exact values of the Fermi wavevector k_F (necessary for the use of the diffraction model) were not known, k_F has been taken as an adjustable parameter. At the same time, if the free-electron model is applicable for the description of transport properties, the exact values of k_F can be deduced from the normal Hall coefficient R_0 . Leaving aside the problem whether the electrons in Ni–metalloid alloys can be described as free ones or not for the later discussion, we thought it interesting to investigate the concentration dependence of R_0 , ρ and α in amorphous Ni–metalloid alloys. In particular, we wished to find out whether a clear correlation between R_0 and ρ or α exists, and furthermore, whether the values for k_F (deduced from R_0) are meaningful within the framework of the diffraction model.

We measured the Hall resistivity (at liquid nitrogen and room temperature), the electrical resistivity and the temperature coefficient of the resistivity (at room temperature) for ten amorphous Ni–B–Si alloys with metalloid concentration ranging from 20 to 39 at.%. The alloys were prepared by melt-spinning technique (Liebermann and Graham 1976). The samples were ribbons about 1.5–2.0 mm wide and on average $24 \mu\text{m}$ thick. The largest error in the Hall resistivity arises from the uncertainty in thickness and

Table 1. Data for Ni–B–Si alloys: the normal Hall coefficient R_0 , the electrical resistivity ρ , the room-temperature coefficient of the resistivity α , the number of the conduction electrons per atom n_a , the Fermi wavevector k_F and the number of electrons that the metalloid atoms possibly contribute to the conduction band n_m .

Alloy	$-R_0$ ($10^{-10} \text{ m}^3 \text{ A}^{-1} \text{ s}^{-1}$)	ρ ($\mu\Omega \text{ cm}$)	α (10^{-4} K^{-1})	n_a	$2k_F$ (10^{-10} m^{-1})	n_m
Ni ₈₀ B ₁₈ Si ₂	1.00	83	1.80	0.65	2.46	0.85
Ni ₈₀ B ₁₅ Si ₅	0.93	87	1.80	0.71	2.52	1.15
Ni ₈₀ B ₁₀ Si ₁₀	0.88	85	1.70	0.76	2.56	1.40
Ni ₇₈ B ₁₂ Si ₁₀	0.74	89	1.35	0.89	2.72	1.92
Ni ₇₅ B ₂₀ Si ₅	0.76	97	1.20	0.82	2.68	1.48
Ni ₇₅ B ₁₅ Si ₁₀	0.70	95	1.10	0.91	2.76	1.84
Ni ₇₅ B ₁₀ Si ₁₅	0.70	110	1.00	0.93	2.76	1.92
Ni ₆₇ B ₃₃	0.50	122	0.22	1.13	3.10	2.21
Ni ₆₇ B ₂₂ Si ₁₁	0.49	138	0.00	1.21	3.12	2.43
Ni ₆₁ B ₃₉	0.45	160	-0.22	1.17	3.20	2.06

it amounts to about 5%. The actual measurements were performed using the standard DC technique.

All our alloys are paramagnetic and the Hall resistivity is a linear function of magnetic field. The values obtained for R_0 , for a particular alloy, at liquid nitrogen and room temperature are (within the experimental error that amounted to about 2%) the same. This indicates that there is no anomalous magnetic contribution to the Hall effect in our alloys.

The values of R_0 , ρ and α for all our NiBSi alloys are listed in table 1. In figure 1 we have plotted the values for α and R_0 as a function of electrical resistivity. As expected,

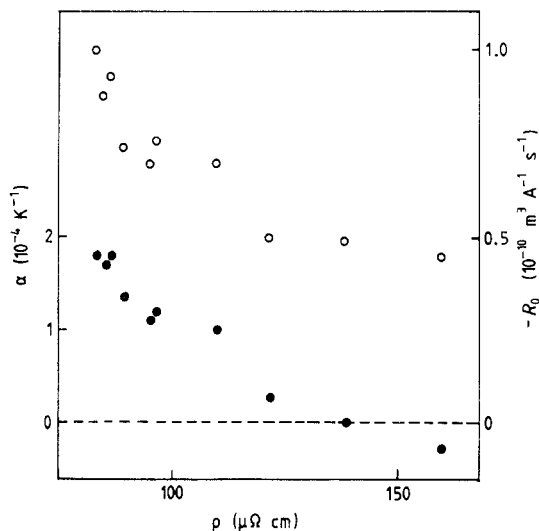


Figure 1. The room-temperature coefficient of the resistivity (●) and the normal Hall coefficient (○) as a function of the electrical resistivity in the Ni–B–Si alloys listed in table 1.

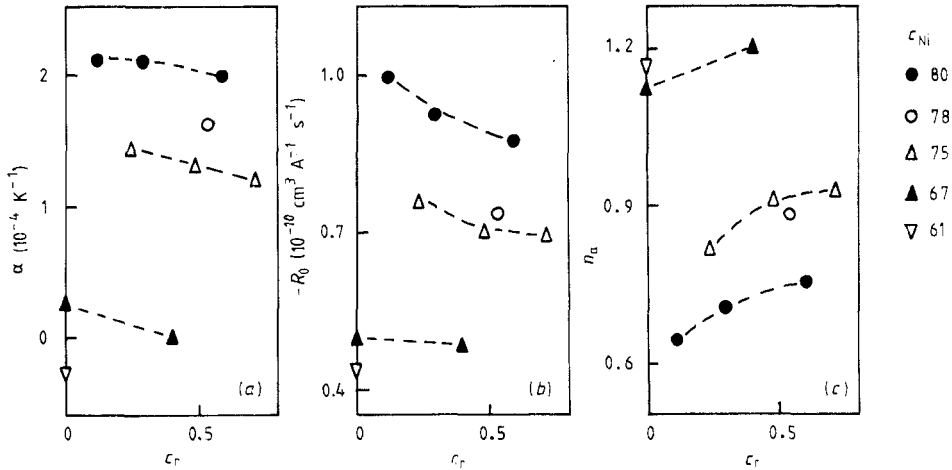


Figure 2. (a) The room-temperature coefficient of the resistivity, (b) the normal Hall coefficient and (c) the number of the conduction electrons per atom in NiBSi alloys as a function of silicon to the total metalloid content ratio c_r . The nickel concentration c_{Ni} is indicated in the figure.

α decreases with increasing resistivity and changes sign for $\rho > 140 \mu\Omega \text{ cm}$. We note that the Hall coefficient also decreases strongly with increasing resistivity.

In order to explore in more detail how the transport properties of our alloys depend on the metalloid concentration, we have plotted, in figure 2(a) and 2(b), α and R_0 as a function of silicon to the total metalloid content ratio. From these figures it can be concluded that, for a fixed silicon to boron ratio, both α and R_0 decrease with increasing total metalloid content and that, for a fixed nickel concentration, both α and R_0 decrease if boron is substituted by silicon. A similar concentration dependence of α in amorphous Ni-B-Si alloys has already been reported by Komatsu *et al* (1987).

Before we proceed to interpret our R_0 data within the framework of the free-electron model, we wish to mention some points that may justify such an interpretation. The principal objection to the use of the Ziman model for the calculation of the transport properties of amorphous or liquid transition metal alloys is its neglect of the d electron contribution to the conductivity (Ballentine 1982, Morgan *et al* 1984). However, in Ni-base amorphous alloys, with metalloid content $\geq 20 \text{ at.}\%$, where the Fermi level E_F is close to the edge of the d band DOS, the electronic transport will be dominated by s-like nearly free electrons and their number density can be deduced from R_0 . Furthermore we note that even in liquid nickel, which certainly has a greater density of d states at E_F than any of our alloys, the value for R_0 (Künzi and Güntherodt 1980) is close to the free-electron value and gives 0.6 electrons per atom in the conduction band, which is equal to the number of holes in the d band.

The data for the number of electrons per atom n_a , calculated in the free-electron model, are listed in table 1. In figure 2(c) we have plotted n_a as a function of silicon to the total metalloid content ratio. We see that, for a fixed silicon to boron ratio, n_a increases with increasing metalloid concentration. At the same time, for a fixed nickel concentration, n_a increases with increasing silicon concentration as would be expected from the valencies of boron and silicon in a simple charge-transfer model.

From the conduction electron density we also calculated the values of the Fermi wavevector $k_F = (3\pi n)^{1/3}$. The data for $2k_F$ are presented in table 1. The concentration dependence of k_F is similar to that of n_a which is plotted in figure 2(c). We note that $2k_F$ increases from $2.46 \times 10^{-10} \text{ m}^{-1}$ in the $\text{Ni}_{80}\text{B}_{18}\text{Si}_2$ alloy to $3.20 \times 10^{-10} \text{ m}^{-1}$ in the $\text{Ni}_{61}\text{B}_{39}$ alloy. At the same time the position of the first peak k_p in the static structure factor $S(k)$ of amorphous Ni-metalloid alloys is practically independent of the concentration in this concentration range (Lampartier *et al* 1982, Gardner *et al* 1985) and amounts to $3.0\text{--}3.2 \times 10^{-10} \text{ m}^{-1}$. From the above considerations we can conclude that the dependence of the resistivity and the dependence of the temperature coefficient of the resistivity on composition in amorphous Ni-B-Si alloys can be interpreted in terms of Ziman-Faber theory. In agreement with this theory the resistivity increases with increasing k_F and a negative temperature coefficient of the resistivity above the Debye temperature occurs when $S(2k_F) > 1$, i.e. when $2k_F$ approaches k_p .

Furthermore, we thought it interesting to estimate what would be the number of electrons (if the simple picture of the charge transfer is assumed) contributed by the metalloid atoms to the conduction band. For this estimate we have assumed that the number of conduction electrons that come from nickel is equal to 0.6 electrons per nickel atom. The number of conduction electrons n_m contributed by the metalloid atoms to the conduction band is listed in table 1. We note that n_m increases significantly with increasing metalloid or increasing silicon concentration. Such a composition dependence of n_m may indicate that as the d band is being filled, more electrons from the metalloid atoms enter the conduction band.

Finally we would like to make some comments on the above, probably oversimplified, interpretation. Recent theoretical work (Khanna *et al* 1985, Press *et al* 1987) emphasises that the electronic structure of Ni-metalloid alloys cannot be properly interpreted in terms of a simple charge transfer and the rigid band model. Instead of this a more accurate picture, which takes into account the hybridisation of the electronic wavefunctions, has to be used. Moreover there are some indications that the s electrons from metalloid atoms do not interact strongly with Ni electrons and instead form localised states with energies below the Ni band. With this in mind, one may object that the values for n_m deduced above are too high. Furthermore, in recent theoretical studies of the Hall effect in amorphous alloys (Movaghar 1985, Nguyen-Manh *et al* 1987), it has been calculated that the Hall coefficient R_0 is proportional to the derivative of the DOS g' at the Fermi level E_F . In the case of strong scattering, i.e. for high resistivities, the DOS at E_F will be lowered and a minimum may even appear. Taking this into account, one can assume that the decrease of R_0 with increasing resistivity reflects the decrease of g' (as for instance is noticed in some liquid metals (Oelhafen *et al* 1988)).

The above findings may apparently make the interpretation of the electronic transport properties of amorphous Ni-B-Si alloys, outlined earlier in this paper, somewhat ambiguous. In particular, although the application of the free-electron model on our experimental R_0 -values yields the k_F -values consistent with the interpretation of the electrical resistivity of Ni-B-Si alloys in terms of the Ziman model, we have no direct proof that this model indeed provides the correct description of the electronic transport in these alloys. Indeed in our alloys the change in sign of α occurs at very high values of ρ , which questions the interpretation of negative α in terms of the Ziman model only. Therefore a detailed study of the temperature and magnetic field dependence of the electrical resistivity (as well as of the other transport properties) is required in order to elucidate the nature of the electronic conduction in amorphous Ni-B-Si alloys. This work is now in progress.

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