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## The Hall effect and the conduction electron density in Ni-based amorphous alloys

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**Abstract.** New results for the normal Hall coefficient  $R_0$ , the resistivity and the room-temperature coefficient of resistivity of five Ni–B–Si and six Ni–P amorphous (melt-quenched) alloys are reported. From these results, and results already reported, we have obtained rather detailed information on the variation in the conduction electron density with the metalloid concentration  $c_m$  in the amorphous Ni–B–Si alloys. The results for the alloys with low  $c_m$  extrapolate to those of liquid Ni, whereas the variation in the Fermi wavevector (deduced from  $R_0$ ) with  $c_m$  supports the applicability of the Ziman–Faber theory for the explanation of the transport properties of these alloys. The contribution of the metalloid atoms to the conduction band is found to change at  $c_m \approx 0.3$ , which is probably related to filling of the d band. This change occurs in the range of  $c_m$  in which a significant change in the chemical short-range order has been observed. Although less detailed, the results for the Ni–P alloys follow the same trend as those for Ni–B–Si alloys and do not indicate any dramatic change in  $R_0$  at around  $c_m \approx 0.24$  as reported for electrodeposited Ni–P alloys.

### 1. Introduction

In a previously published paper on the Hall effect and the electrical resistivity in amorphous Ni–B–Si alloys [1] (hereafter referred to as I) we have concluded that the values of the Fermi wavevector  $k_F$ , deduced from the normal Hall coefficient  $R_0$ , are consistent with the interpretation of the electronic transport in Ni–metalloid metallic glasses in terms of the Ziman–Faber theory [2, 3]. It has also been concluded, from the composition dependence of the conduction electron density, that the number of electrons contributed by the metalloid atoms to the conduction band increases significantly with increasing metalloid or increasing silicon concentration.

More recently Očko and Babić [4] have obtained mutually consistent results for the resistivity  $\rho$ , the temperature coefficient  $\alpha$  of resistivity at room temperature and the thermoelectric power in amorphous Ni–P–B alloys within the framework of the extended Ziman model. In their calculations they have assumed that the number of d electrons and the number of conduction electrons are specific functions of boron and phosphorus concentrations.

Therefore, we have decided to find out whether some more quantitative results concerning the particular contributions of the metalloid atoms to the conduction band could be deduced from the Hall effect measurements. We also intend to compare the

electronic transport properties in amorphous Ni–P and Ni–B–Si alloys. For this purpose, we have measured the Hall effect and the electrical resistivity in six  $\text{Ni}_{100-x}\text{P}_x$  amorphous alloys ( $17 \leq x \leq 22$ ) and completed the results from I with measurements on five additional Ni–B–Si alloys with the total metalloid concentration close to 30 at. %.

## 2. Experimental procedure

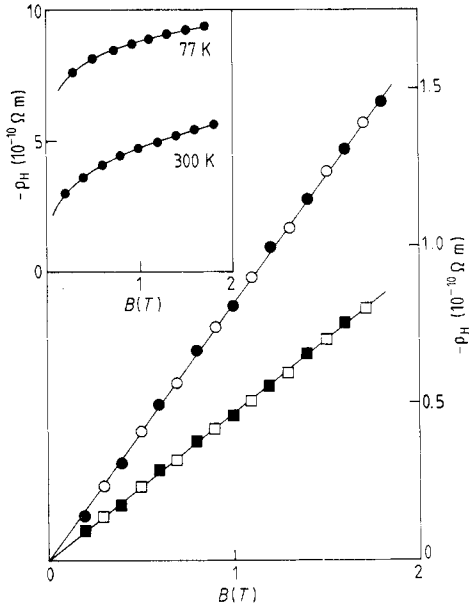
The amorphous alloys were prepared by the melt-spinning technique [5]. We note here that Ni–P alloys with a phosphorus concentration smaller than 17 at. % or greater than 22 at. % are hard to obtain in an amorphous state. The samples were ribbons about 1.5–2.5 mm wide and on average 25  $\mu\text{m}$  thick. The thickness was determined from mass, density, length and width measurements. The uncertainty in thickness is the largest source of error in the absolute Hall resistivity results which are accurate to within 5%. The samples were cut into a standard shape used for the Hall measurements. The Hall resistivity was measured by the five-probe method and by standard DC and AC techniques with a relative accuracy equal to about 0.5%. The Hall voltage leads (50  $\mu\text{m}$  Cu wires) were soldered onto the samples with a low-temperature solder. In the case of  $\text{Ni}_{83}\text{P}_{17}$  alloy, where it is possible that soldering induces the formation of the ferromagnetic phase [6], the measurements were repeated with the contacts glued with a silver paint and the results were found to be the same as obtained with the soldered contacts.

The measurements were performed in magnetic fields up to 2 T. The Hall voltages were measured at room temperature and liquid-nitrogen temperature. The resistivity measurements were performed in the temperature range between 273 and 320 K with the standard four-probe method. Either an AC or a DC technique was employed and the current passing through the samples was 10 mA or less.

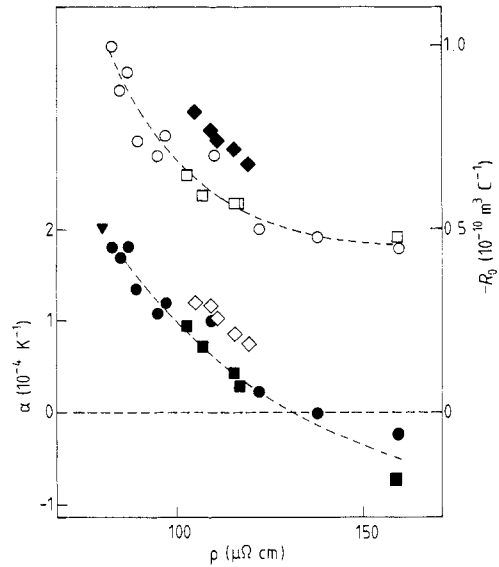
## 3. Results and discussion

The Hall resistivity  $\rho_{\text{H}}$  as a function of magnetic field at 300 and 77 K for three of our alloys is shown in figure 1. In the case of the  $\text{Ni}_{83}\text{P}_{17}$  alloy, a term non-linear in the field, which strongly depends on temperature, is observed. It is evident that this alloy either is ferromagnetic or contains a ferromagnetic phase with a Curie temperature of around room temperature. Because of this, in our discussion we shall not further consider the results obtained for this particular alloy. We wish only to point out that, in amorphous Ni–P alloys, ferromagnetism disappears at about 17 at. % P [7]. In all our other alloys with a higher metalloid concentration,  $\rho_{\text{H}}$  was, as expected, a linear function of the magnetic field and was almost independent of temperature ( $T > 77$  K). This indicates that there is no anomalous magnetic contribution to the Hall effect in these alloys.

The values of  $R_0$ ,  $\rho$  and  $\alpha$  for paramagnetic Ni–P and Ni–B–Si alloys investigated in the present work are listed in table 1. In figure 2 we have plotted the present values of  $\alpha$  and  $R_0$  together with the corresponding data from I as functions of electrical resistivity. We also included in figure 2 the result for the resistivity and the temperature coefficient of resistivity for liquid nickel extrapolated to the room temperature (to be referred to, for simplicity, as  $\rho$  and  $\alpha$  of liquid nickel). In this extrapolation the resistivity of liquid nickel ( $\rho \approx 90 \mu\Omega \text{ cm}$  at 1600 °C) was scaled down to that at 300 K by taking into account the measured  $d\rho/dT$  ( $\approx 0.015 \mu\Omega \text{ cm K}^{-1}$  at 1800 °C) in the liquid state [8]. We note that the data for  $\rho$  and  $\alpha$  for liquid nickel are close to those for Ni–B–Si alloys with the lowest



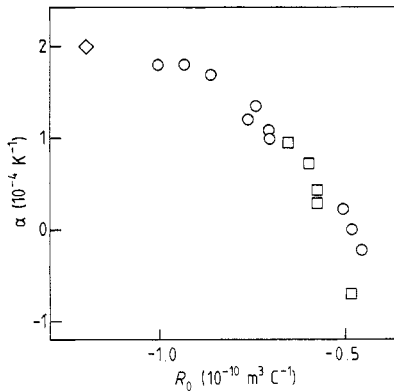
**Figure 1.** The Hall resistivity as a function of magnetic field for  $\text{Ni}_{82}\text{P}_{18}$  ( $\circ$ ,  $\bullet$ ) and  $\text{Ni}_{65}\text{B}_{20}\text{Si}_{15}$  ( $\square$ ,  $\blacksquare$ ) alloys at room temperature ( $\bullet$ ,  $\blacksquare$ ) and liquid-nitrogen temperature ( $\circ$ ,  $\square$ ). The inset shows  $\rho_H$  versus the magnetic field for  $\text{Ni}_{83}\text{P}_{17}$  alloy at 300 and 77 K.



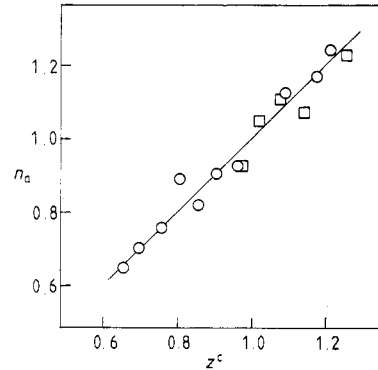
**Figure 2.** The normal Hall coefficient and the room-temperature coefficient of resistivity as a function of the electrical resistivity:  $\square$ ,  $R_0$  for Ni-B-Si alloys from table 1;  $\blacklozenge$ ,  $R_0$  for Ni-P alloys from table 1;  $\circ$ ,  $R_0$  for Ni-B-Si alloys from I;  $\blacksquare$ ,  $\alpha$  for Ni-B-Si alloys from table 1;  $\diamond$ ,  $\alpha$  for Ni-P alloys from table 1;  $\bullet$ ,  $\alpha$  for Ni-B-Si alloys from I;  $\blacktriangledown$ ,  $\alpha$  and  $\rho$  extrapolated from the liquid-nickel values [8].

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

Alloy	$-R_0$ ( $10^{-10} \text{ m}^3 \text{ C}^{-1}$ )	$\rho$ ( $\mu\Omega \text{ cm}$ )	$\alpha$ ( $10^{-4} \text{ K}^{-1}$ )	$n_a$	$2k_F$ ( $10^{10} \text{ m}^{-1}$ )	$n_m$
$\text{Ni}_{72}\text{B}_{26}\text{Si}_2$	0.65	103	0.93	0.93	2.82	1.78
$\text{Ni}_{72}\text{B}_{20}\text{Si}_8$	0.59	107	0.70	1.05	2.92	2.20
$\text{Ni}_{72}\text{B}_{14}\text{Si}_{14}$	0.57	116	0.42	1.11	2.96	2.42
$\text{Ni}_{70}\text{B}_{20}\text{Si}_{10}$	0.57	117	0.29	1.07	2.96	2.16
$\text{Ni}_{65}\text{B}_{20}\text{Si}_{15}$	0.48	159	-0.70	1.23	3.14	2.40
$\text{Ni}_{82}\text{P}_{18}$	0.82	105	1.20	0.83	2.62	2.00
$\text{Ni}_{81}\text{P}_{19}$	0.77	109	1.17	0.90	2.68	2.19
$\text{Ni}_{80}\text{P}_{20}$	0.75	111	1.02	0.92	2.70	2.21
$\text{Ni}_{79}\text{P}_{21}$	0.72	116	0.85	0.96	2.74	2.29
$\text{Ni}_{78}\text{P}_{22}$	0.68	116	0.76	1.01	2.78	2.45



**Figure 3.** The room-temperature coefficient of the resistivity as a function of the normal Hall coefficient in amorphous Ni-B-Si alloys from table 1 ( $\square$ ) and from I ( $\circ$ ):  $\diamond$ ,  $R_0$  [10] and  $\alpha$  for liquid nickel.



**Figure 4.** The number  $n_a$  of conduction electrons determined from  $R_0$  versus the calculated number  $z^c$  of conduction electrons for the Ni-B-Si alloys from this work ( $\square$ ) and from I ( $\circ$ ).

metalloid content (I).

As has already been reported in I, both  $R_0$  and  $\alpha$  decrease with increase in resistivity. In Ni-B-Si alloys,  $\alpha$  changes sign, as one would expect from the Mooij [9] correlation, for a resistivity close to  $140 \mu\Omega \text{ cm}$ . Concerning the dependence of  $R_0$  and  $\alpha$  on the alloy composition, we note that in general both  $R_0$  and  $\alpha$  decrease with increasing metalloid content. In Ni-B-Si alloys both  $R_0$  and  $\alpha$  decrease if boron is substituted by silicon. At the same time in Ni-P alloys both  $R_0$  and  $\alpha$  are lower than in Ni-B-Si alloys for the same total metalloid concentration.

In figure 3 we have plotted the room-temperature coefficient of the resistivity as a function of the normal Hall coefficient for all Ni-B-Si alloys. The corresponding results for Ni-P alloys will be discussed later. In figure 3 we also included the data for  $R_0$  [10] and  $\alpha$  of liquid nickel which fit well to the curve defined by  $\alpha$ - and  $R_0$ -values of amorphous Ni-B-Si alloys. From figures 2 and 3 we can conclude that, while  $\alpha$  strongly decreases and changes its sign with increase in  $c_m$ , the normal Hall coefficient slows down its decrease with increasing metalloid concentration. There are no indications that  $R_0$  would change its sign and become positive on further increase in the metalloid content. In our opinion the decrease in  $R_0$  with increase in metalloid (or silicon) content in the Ni-B-Si alloys simply reflects the increase in the conduction electron density upon alloying. This point will be discussed in some detail later on.

In the interpretation of  $R_0$  data we follow the same arguments as those used in I. First, we neglect the d-band contribution to the conduction electron density and then we treat the conduction electrons as free electrons. These two approximations are generally accepted in the interpretation of the electronic transport properties of disordered alloys provided that the d-band density of states at the Fermi level and the resistivity are not too high [11, 12]. We think that these conditions are fulfilled in the case of our alloys, although for the higher metalloid concentrations the resistivity approaches the strong scattering region.

From the values of  $R_0$ , i.e. from the conduction electron density, we have calculated the values of the Fermi wavevector  $k_F$ . The data for  $2k_F$  are presented in table 1 and they complete the previously reported results in I. Now, for completeness, we shall briefly review the main conclusions of I.

According to the diffraction model, i.e. the Ziman–Faber theory, the resistivity in disordered alloys increases by increasing  $k_F$ , and a negative temperature coefficient of the resistivity above the Debye temperature occurs when  $2k_F$  approaches the value  $k_p$  for which the first peak in the static structure factor  $S(k)$  occurs. The value of  $k_p$  in Ni–metalloid amorphous alloys is, in the concentration range under consideration, almost independent of the composition of the alloy and equals  $(3.0\text{--}3.2) \times 10^{10} \text{ m}^{-1}$  [13, 14]. At the same time in amorphous Ni–B–Si alloys,  $2k_F$  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. From this we have concluded that the dependence of  $\alpha$  and  $k_F$  on the alloy composition justifies the interpretation of the electronic transport in Ni–metalloid amorphous alloys in terms of the Ziman–Faber theory. Here we note that the values of  $k_F$  deduced from  $R_0$  in our Ni–metalloid alloys are close to those used by Očko and Babić [4] in the calculations of the electronic transport coefficients in a similar alloy system. For example, the value of  $k_F$  in  $\text{Ni}_{82}\text{P}_{18}$  alloy (table 1) agrees well with the value of  $k_F$  used in their calculations dealing with  $\text{Ni}_{81.5}\text{P}_{18.5}$  alloy.

Now we estimate the number  $n_m$  of electrons contributed by the metalloid atoms to the conduction band. For this estimation we have assumed that the number of conduction electrons that come from nickel is equal to 0.6 electrons per nickel atom (as can be concluded from  $R_0$  of liquid nickel). The values of  $n_m$  are given in table 1 of this paper and in I as well. In general we conclude that  $n_m$  increases with increasing metalloid concentration  $c_m$  or, in the case of Ni–B–Si alloys, with increasing silicon concentration. At the same time,  $n_m$  is larger in Ni–P alloys than in Ni–B–Si alloys of the same total metalloid content. As the determination of the exact influence of the metalloid concentration on the electronic properties of Ni-based alloys is of considerable interest, we decided to determine a more quantitative relation that would separate the boron and silicon contributions to the conduction band.

On the basis of the results for the alloys with a low silicon concentration presented here and in I we made a rough estimation that the number of boron electrons contributing to the conduction band increases linearly with  $c_m$  from 0.8 electrons per boron atom for  $c_m = 0.2$  to 2.1 electrons per boron atom for  $c_m = 0.3$ . In this case, in order to fit the data, the number of electrons contributed by silicon should increase linearly with  $c_m$  from 2 to 3 electrons per silicon atom in the same range of total metalloid content. For  $c_m > 0.3$  the numbers of electrons contributed by boron and silicon atoms are roughly constant and equal to 2.1 and 3, respectively. Under these assumptions we have calculated the number  $z^c$  of conduction electrons according to the following relations:

$$z^c = 0.6c_{\text{Ni}} + [0.8 + 13(c_m - 0.2)]c_{\text{B}} + 10c_m c_{\text{Si}} \quad (1)$$

for  $0.2 \leq c_m \leq 0.3$ , and

$$z^c = 0.6c_{\text{Ni}} + 2.1c_{\text{B}} + 3c_{\text{Si}} \quad (2)$$

for  $c_m > 0.3$ . Here,  $c_{\text{Ni}}$ ,  $c_{\text{B}}$  and  $c_{\text{Si}}$  are the concentrations of nickel, boron and silicon, respectively. We note that the recent measurements of the thermoelectric power in the same Ni–B–Si alloys [15] show that the dependence of the thermopower on the alloy composition changes significantly at the metalloid concentrations close to 30 at.% as well.

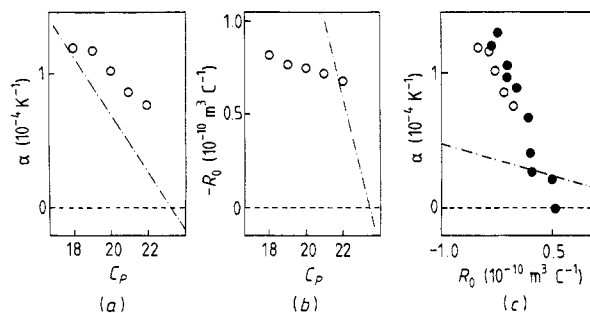
In figure 4 we have plotted the number  $n_a$  of conduction electrons per atom deduced from  $R_0$  versus the calculated number  $z^c$ . In our opinion, the agreement between  $n_a$  and  $z^c$  is very good and justifies the assumptions involved in the calculation of  $z^c$ . (Among these assumptions, the fact that the Ni contribution does not change upon alloying,

although commonly used when dealing with the transition-metal-based alloys, is probably the most arbitrary.)

Now we discuss some possibilities that may produce such a composition dependence of  $n_a$ . Within the framework of the rigid-band model such a dependence of  $z^c$ , i.e. of  $n_a$  on the metalloid content in Ni–B–Si alloys would reflect the filling of the d band and the decrease in the d-band density of states in these alloys. For  $c_m > 0.3$  the d band is probably filled and the metalloid contribution to  $n_a$  is roughly constant. In the range  $0.2 \leq c_m \leq 0.3$ , the d-band density of states decreases while the number of electrons that metalloid atoms contribute to the conduction band increases with increasing metalloid concentration. However, the rigid-band model (although often very useful) gives a somewhat oversimplified interpretation of the alloy properties. A more accurate picture should take into account the actual hybridization of the electronic wavefunctions [16, 17]. From this standpoint, we note that the local environment around the metalloid atoms can influence the conduction electron density as well. It is well known that the local environment has a considerable influence on the magnetic properties of transition-metal–metalloid amorphous alloys [18]. Concerning the local environment around the metalloid atoms, it is established that in transition-metal–metalloid amorphous alloys, for the metalloid concentrations near 20 at. % there are no metalloid–metalloid nearest neighbours [19]. On the other hand, it has been reported that in Ni<sub>64</sub>B<sub>36</sub> amorphous alloy a boron atom has on average approximately 1.1 nearest-neighbour boron atoms [13]. From this it seems certain that the local environment around the metalloid atoms changes significantly within the interval of concentrations studied in our alloy system. Therefore, a more detailed explanation of the electronic properties of the Ni–metalloid amorphous alloys should also take into account the effects of the chemical short-range order. The work in which we wish to study the interplay between the local environment effects and the d-band filling in these alloys is in progress now.

Finally, we shall briefly discuss the results for the Ni–P alloys, especially because the results for the Hall effect, which are different from those presented in this work, have been reported for the electrodeposited Ni–P alloys [20]. In the electrodeposited Ni–P amorphous alloys the Hall coefficient strongly depends on the alloy composition and with increasing phosphorus concentration changes sign, from a negative to a positive value. In order to compare the results for melt-quenched and electrodeposited alloys, we have plotted in figures 5(a) and 5(b) the concentration dependence of  $\alpha$  and  $R_0$  in those alloys, respectively. Whereas the dependence of  $\alpha$  on the phosphorus concentration is similar in the alloys prepared by different techniques (i.e.  $\alpha$  decreases at a similar rate in both systems), the concentration dependence of  $R_0$  is quite different.  $R_0$  in melt-quenched Ni–P alloys decreases slowly with increase in the phosphorus concentration but it does not show any marked tendency to change sign as it does in electrodeposited alloys. In figure 5(c) we have plotted  $\alpha$  as a function of  $R_0$  for melt-quenched Ni–P and Ni–B–Si alloys and for electrodeposited Ni–P alloys. It can be seen that the correlation between  $\alpha$  and  $R_0$  in melt-quenched Ni–P alloys is very similar to that for melt-quenched Ni–B–Si alloys. Therefore, the electronic properties of the melt-quenched Ni–metalloid amorphous alloy appear to be qualitatively similar, regardless of the actual type of metalloid involved. The differences between the particular melt-spun alloys can be simply related to the composition of the alloys and to the valences of their ingredients.

The origin of the different behaviour of the Hall coefficient in the electrodeposited alloys is not at present quite clear to us. However, we note that it has been reported that, in non-ferromagnetic electrodeposited Ni–P alloys, chemical inhomogeneities or clusters exist [7]. The nickel concentration of these clusters increases with increasing



**Figure 5.** (a) The room-temperature coefficient of the resistivity in melt-quenched (○) and electrodeposited (— · —, line from figure 1 of [21]) Ni-P alloys as a function of the phosphorus concentration. (b) The normal Hall coefficient in melt-quenched (○) and electrodeposited (— · —, data for four non-ferromagnetic samples with 18 at. %  $\leq C_p \leq 26$  at. % from [20]) Ni-P alloys as a function of the phosphorus concentration. (c)  $\alpha$  as a function of  $R_0$  in melt-quenched Ni-P (○) and Ni-B-Si (●) alloys and in electrodeposited (— · —) Ni-P alloys.

phosphorus concentration of the alloy [7]. Further, a positive sign of the anomalous Hall effect (contrary to the negative anomalous Hall effect in crystalline nickel) has been observed in some amorphous Ni-metalloid alloys [22]. At the same time, the Hall coefficient in the electrodeposited Ni-P alloys has been determined at room temperature only. On the basis of the factors mentioned above, we conclude that there is the possibility that the magnetic state of the samples affects the Hall effect measurements in these alloys. This may be the origin of the difference between our  $R_0$ -values for the melt-spun alloys and the Hall coefficients of the electrodeposited alloys [20]. This would be in agreement with the fact that the Hall effect (figure 5(b)) is much more sensitive to the magnetic state of the sample than the resistivity is (figure 5(a)).

#### 4. Conclusion

We measured  $\rho$ ,  $\alpha$  and  $R_0$  in the melt-quenched Ni-B-Si and Ni-P alloys. In general both  $R_0$  and  $\alpha$  decrease with increasing metalloid content. In the Ni-B-Si alloys,  $R_0$  and  $\alpha$  decrease if boron is substituted by silicon and, in the Ni-P alloys,  $R_0$  and  $\alpha$  are lower than in the Ni-B-Si alloys with the same total metalloid content. The values of the Fermi wavevector  $k_F$  deduced from  $R_0$  using the free-electron model are consistent with the interpretation of the electronic transport in Ni-B-Si alloys within the framework of the Ziman-Faber theory.

The number of the conduction electrons in the Ni-B-Si alloys can be calculated from the alloy composition provided that the plausible assumptions concerning the number of electrons that boron and silicon contribute to the conduction band are made. According to these assumptions, the number of electrons contributed to the conduction band by boron and silicon increases from 0.8 to 2.1 electrons per boron atom and from 2 to 3 electrons per silicon atom as the total metalloid content increases from 20 to 30 at. %. In the alloys of the higher metalloid content the number of electrons contributed by boron and silicon is roughly independent of the alloy composition and is equal to 2.1 and 3 electrons per boron and silicon atom, respectively. Such a dependence of the number of electrons contributed to the conduction band by the metalloid atoms would, within the rigid-band model, reflect the filling of the d band upon alloying. It is interesting to



note that a filling of the d band in Ni–B–Si alloys seems to be completed in the range of metalloid concentrations in which a significant change in the local environment around the metalloid atoms is expected [13]. In order to study in some detail the interplay between the d-band filling and the local environment effects, the measurements on Ni–metalloid alloys with higher concentrations of some other metalloid are required.

Concerning the normal Hall coefficient  $R_0$  in disordered Ni–metalloid alloys with the metalloid content lower than in alloys studied here and in I, we expect that it will, with decrease in the metalloid concentration, slowly approach the value of  $-1.2 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ , i.e. the value of  $R_0$  in liquid nickel. However, the amorphous structure of Ni–metalloid alloys with metalloid content lower than about 20 at. % is in general less stable. Even if this structure were stable, as in the  $\text{Ni}_{83}\text{P}_{17}$  alloy, it is evident that ferromagnetic effects would appear. The anomalous Hall effect will then make the proper determination of  $R_0$  a delicate task, as is usual in the case of disordered ferromagnetic alloys [23]. In spite of this, we think that it is worthwhile to extend the measurements reported here by the investigations of the electron transport in disordered Ni alloys (perhaps paramagnetic liquid alloys) with a lower metalloid concentration. Such measurements may provide a better insight into the extent of the applicability of the Faber–Ziman theory to the disordered Ni-based alloys.

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