## **Transport properties and point-contact spectra of**  $N_i/N_{1-x}$  **metallic glasses**

A. Halbritter

*Department of Physics, Institute of Physics, Technical University of Budapest, 1111 Budapest, Hungary*

O. Yu. Kolesnychenko

*Research Institute for Materials, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands*

G. Mihály

*Department of Physics, Institute of Physics, Technical University of Budapest, 1111 Budapest, Hungary*

O. I. Shklyarevskii

*Research Institute for Materials, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands*

*and B. Verkin Institute for Low Temperature Physics & Engineering, National Academy of Science of Ukraine, 47 Lenin Avenue, 310 164 Kharkov, Ukraine*

H. van Kempen

*Research Institute for Materials, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands* (Received 8 June 1999)

Bulk resistivity and point contact spectra of  $N_i_N N_{1-x}$  metallic glasses have been investigated as functions of temperature  $(0.3-300 \text{ K})$  and magnetic field  $(0-12 \text{ T})$ . Metallic glasses in this family undergo a superconducting phase transition determined by the Nb concentration. When superconductivity was suppressed by a strong magnetic field, both the bulk sample  $R(T)$  and the point contact differential resistance curves of  $Ni<sub>x</sub>Nb<sub>1-x</sub>$  showed logarithmic behavior at low energies, which is explained by a strong electron-'two level system'' coupling. We studied the temperature, magnetic field, and contact resistance dependence of  $Ni_{44}Nb_{56}$ point-contact spectra in the superconducting state and found telegraphlike fluctuations superimposed on superconducting characteristics. These  $R(V)$  characteristics are extremely sensitive detectors for slow relaxing "two level system'' motion.

Amorphous metallic alloys have been the subject of extensive investigations over the last two decades because of their surprising transport, magnetic, and superconducting properties. To explain the most unusual low-temperature physical properties of glassy materials the concept of twolevel tunneling systems (TLS) was suggested (for a review see Ref. 1). According to a theoretical model, $^{2}$  the electron scattering processes in TLS result in the commonly occurring logarithmic temperature dependence of the resistivity. On the other hand the resistivities of some amorphous alloys  $(Ni_xNb_{1-x}, Fe_xAu_{1-x})$  were claimed not to exhibit any logarithmic feature at low temperatures<sup>3</sup> and therefore these alloys were regarded as a different class of metallic glasses.

Point contact  $(PC)$  spectroscopy<sup>4</sup> offers a very sensitive method to investigate scattering processes in conducting materials, since back scattering of electrons in a PC causes a noticeable change in the current through the PC and the *R*(*V*) characteristics give quantitative information about the energy dependence of the electron scattering processes on quasiparticles (TLS, phonons, magnons, etc.). In particular, the strong electron-TLS coupling gives rise to a peak in the PC differential resistance around  $V=0$ . This phenomenon is usually referred to as the zero bias anomaly (ZBA). Experiments on metallic PC's containing nonequilibrium defects<sup>5,6</sup> as well as on metallic glass PC's (Refs. 7,8) demonstrated the existence of ZBA suggested by the theory. $2.9$ 

The present study was motivated by the expected absence of the logarithmic peak in  $Ni_xNb_{1-x}$ , and was aimed at investigating TLS scattering in these metallic alloys with the help of PC spectroscopy. The  $Ni_xNb_{1-x}$  metallic glasses  $(MG's)$  are also interesting from another point of view, since a superconducting ground state develops at high enough Nb concentration.<sup>10</sup> We examined  $Ni_{44}Nb_{56}$  and  $Ni_{59}Nb_{41}$  by measurements on bulk samples investigating the temperature and magnetic field dependence of resistivity and by PC spectroscopy based on break junction technique.<sup>11</sup> This technique permits very stable PC in the resistance range  $1-200 \Omega$  to be made by breaking the sample in ultrahigh vacuum and then forming the contact between the freshly fractured atomically clean surfaces. Due to the relatively large resistivity of  $Ni<sub>x</sub>Nb<sub>1-x</sub> MG$  these contacts were basically in the Maxwell limit,<sup>4</sup> where the contact resistance is calculated as  $R_{\text{PC}}$  $= \rho/d$  ( $\rho$  being the electrical resistivity, *d* the contact diameter!.

Figure 1 presents the results of bulk sample measurements. We found that  $Ni_{44}Nb_{56}$  has a superconducting transition at 1.8 K, while the resistance of  $Ni_{59}Nb_{41}$  starts to decrease only below 700 mK indicating a superconducting transition just outside of the temperature range of the measurements (300 mK). For  $Ni<sub>59</sub>Nb<sub>41</sub>$  we observed logarithmic behavior between 700 mK and 25 K. $^{12}$  It is hard to confirm the logarithmic character for  $Ni_{44}Nb_{56}$  because of the interfering presence of the superconducting fluctuations. In our experiments these fluctuations start at 8 K, where the *R*(*T*) curve splits from that of the nonsuperconducting sample.



FIG. 1. Temperature dependence of resistance in bulk  $Ni_rNb_{1-r}$ samples at  $B=0$  and 12 T.  $\Delta R/R$  represents the relative change of resistance normalized to the 50 K value. The zero field  $(1)$  and 12 T (2) measurements are shown with crosses for  $Ni_{44}Nb_{56}$  and dots for  $Ni_{59}Nb_{41}$ . The inset shows the magnetic field dependence of resistance in the superconducting sample at  $T = 300$  mK.

This broad range of fluctuations is in good quantitative agreement with theoretical calculations<sup>13</sup> and experiments on other amorphous superconductors.<sup>14</sup> The inset in Fig. 1 shows a similarly broad magnetic field region of superconducting fluctuations  $(1.5–2.6 \text{ T})$  at constant temperature  $(300$ mK). After suppressing the superconductivity by applying a magnetic field of 12 T the temperature dependence of the resistivity in both samples shows a clear logarithmic behavior up to 25 K. In this logarithmic region a small but noticable magnetic resistance is observed. Above the temperature range shown in Fig. 1 both samples exhibited decreasing resistivity with increasing temperature. The normal state resistivity at 50 K was  $\rho \approx 2.5 \mu \Omega$  m for Ni<sub>44</sub>Nb<sub>56</sub> and  $\rho$  $\approx$  1.6  $\mu\Omega$  m for Ni<sub>59</sub>Nb<sub>41</sub>.

The PC spectra measurements showed a common behavior of  $dV/dI(V)$  for all contacts in the resistance range  $R_{PC}$  $=1-30 \Omega$  (corresponding to the contact diameter *d*  $=$  2000–60 nm) which means rather good reproducibility of results for different samples. For  $R_{PC}$ >60  $\Omega$  (*d*< 30 nm) individual features start to prevail and the *dV*/*dI*(*V*) curves for the samples of the same resistance may differ significantly. This sets the length scale of the material inhomogeneity to  $\sim$  30 nm. This value is in agreement with the small angle neutron scattering measurements performed on these metallic glasses $15$  where inhomogeneity was found on the length scale of  $\sim$  18 nm. The regime of electron flow in point contacts depends on the relationship between the contact diameter *d* and the elastic ( $l_{el}$ ) and the inelastic ( $l_{in}$ ) mean free paths.<sup>4</sup> Due to the large resistivity of  $Ni<sub>x</sub>Nb<sub>1-x</sub>MG$ , the transition to the thermal limit  $(l_{el}, l_{in} \le d)$  for low ohmic contacts occurs at small voltage bias (because of the strong energy dependence of the inelastic mean free path). In the thermal regime the excess electron energy is dissipated inside the contact, which results in the increase of the point contact temperature with respect to the bias voltage<sup>4</sup>  $T_{\text{PC}}^2 = T_{\text{bath}}^2$  $V^2/4L$ , where *L* is the Lorenz number. This equation relates  $R(V)$  measurements done by PC technique to the temperature dependence of the resistivity.

Figure 2 shows the differential resistance of  $Ni_{59}Nb_{41}$ junctions. The low ohmic contacts  $(curve 1)$  exhibit clear logarithmic peaks in the voltage region of 1–12 mV, as pre-



FIG. 2. Point contact differential resistance curves of  $Ni<sub>59</sub>Nb<sub>41</sub>$ sample at  $T=1.2$  K. Curve 1 with the right scale represents the logarithmic zero bias anomaly in a low ohmic contact  $(4 \Omega)$ ; curve 2 with the left scale shows the reproducing high-bias singularities in a high ohmic contact (64  $\Omega$ ). The relative change of the differential resistance is normalized to the  $V = 50$  mV value.

sented on the enlarged scale for a 4  $\Omega$  junction. According to the above equation and calculating by the standard Lorenz number, the bias voltage 12 meV corresponds to  $T_{PC}$  $=$  38 K in the thermal regime, which is somewhat higher than the border of the logarithmic region  $(25 K)$  in the bulk sample measurements of  $R(T)$ . This difference is due to the high resistivity of the material: the voltage drop in the vicinity of the contact is comparable with the voltage drop over the bulk part of the sample, which shifts the logarithmic region towards higher voltages. The decrease in the contact resistance between 0 and 50 mV is comparable to that for the bulk samples in the temperature range 5–160 K.

The  $dV/dI(V)$  dependences for high ohmic  $Ni_{59}Nb_{41}$  $\mu$  junctions show steplike singularities at high biases (curve 2, Fig. 2), which can be repeatedly reproduced for the same contact but vary in amplitude and position for different samples. The origin of these high bias anomalies is the subject of ongoing investigations.

The PC characteristics of the superconducting  $Ni_{44}Nb_{56}$ MG below the critical temperature are quite different from these in ordinary superconductors and can be understood only qualitatively.

Figure 3 shows the PC differential resistance and *I*-*V* curves of  $Ni_{44}Nb_{56}$  at different contact resistances. The junc-



FIG. 3. Point contact characteristics of the superconducting  $Ni<sub>44</sub>Nb<sub>56</sub>$  sample at different normal state resistances. (a) Differential resistance curves for  $R_N = 2 - 16 \Omega$  contacts. (b) *I-V* curves of  $R_N$ =0.5,1,2,4,8,16  $\Omega$  contacts, respectively.



FIG. 4. The *I-V* curves of a 1  $\Omega$  Ni<sub>44</sub>Nb<sub>56</sub> point contact in different magnetic fields, at 1.2 K. The inset shows the differential resistance curve of a 11.5  $\Omega$  contact in *B* = 5 T magnetic field. The logarithmic ZBA is regained if superconductivity is totally suppressed.

tions with small normal-state resistance ( $\leq 1.5 \Omega$ ) present conventional *I*-*V* curves [Fig. 3(b)] of a current driven contact with a clear voltage jump above a certain critical current value and with excess current at high voltages. The evaluation of the excess current<sup>16</sup> for these low resistance junctions shows, that the normal resistance — excess current product is constant giving the close-to-BCS value of  $3.2 \pm 0.2$  for  $2\Delta/k_B T_c$ . For higher resistances the *I*-*V* curves are smeared,  $R_N I_{\text{exc}}$  vanishes and an increasing residual resistance is observed at zero bias. We found that this residual resistance increases rapidly for decreasing contact diameter and may differ significantly for contacts with the same  $R_N$ . The transition between the jumplike curves and the smeared ones is also sample dependent, varying between  $1-2 \Omega$ . These phenomena can be understood qualitatively in terms of percolationlike superconductivity. In large enough contacts the current can find continuous superconducting percolation paths between the two electrodes, but below a certain contact diameter no such paths exist any more, and the current must flow through normal regions as well. In this case the residual resistance is determined by the fraction of normal and SC regions along the current paths, which explains the strongly contact-dependent behavior. The characteristic width of percolation paths is most probably close to the material inhomogeneity scale of 18 nm.<sup>15</sup> This size scale of percolation is in agreement with the value of coherence length ( $\approx$  10 nm) calculated from  $H_{c2}$ .

As Fig. 4 shows, the steplike *I*-*V* curve is smeared under the influence of magnetic field as well, but the zero bias resistance and the excess current remains constant up to 1.2 T. We believe that this smearing follows from the vortex dynamics at high current densities in the contact area: the resistance caused by vortices is superimposed on the steplike zero-field *I*-*V* curve. The  $B=1.6$  and 2 T curves are already within the fluctuation region of  $H_c$ <sub>2</sub> (see inset in Fig. 1), thus one obtains completely different *I*-*V* curves with larger zero bias resistance and small excess current. Going above  $H_{c2}$ , at  $B=5$  T we regain the positive logarithmic peak attributed to electron scattering on two level systems (see inset in Fig. 4).

Recording the differential resistance curves of  $Ni_{44}Nb_{56}$ as the function of temperature, we found that the transition is



FIG. 5. The telegraph noise superimposed on the superconducting characteristic of a 15  $\Omega$  Ni<sub>44</sub>Nb<sub>56</sub> contact at the temperature of 1.2 K. The inset represents the zero bias resistance switching between two discrete states as the function of time.

broadening by decreasing contact diameter. Similar behavior was observed by Naidyuk *et al.* in superconducting heavy fermion point contacts.<sup>17</sup>

The differential resistance of some contacts displays large fluctuations around zero bias  $(Fig. 5)$ . The plot of this noise as the function of time shows that the resistance is switching between two (or more) discrete states on the time scale of seconds. This slow two level fluctuation is not sensitive to magnetic fields up to 1.5 T and decreases rapidly at larger fields. In Ref. 7 a similar fluctuation was superimposed on the logarithmic ZBA in  $Fe_{80}B_{20}$  and  $Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$  metallic glasses. This fluctuation was explained as the effect of slowly moving defects influencing electron-TLS coupling. In  $Ni_{44}Nb_{56}$  contacts the motion of such relatively large defects can result in shutting down one of the percolation paths and suppressing superconductivity in a sizeable part of the constriction. These fluctuations were only observed in relatively small contacts ( $d \le 200$  nm). In such small areas only a few percolation paths are present, which explains that shutting down one of them has an observable effect. It makes the superconducting characteristics an extremely sensitive detector for the slow relaxing TLS motion.

In conclusion we demonstrated that in contrast to earlier observations both the bulk resistivity and the PC differential resistance of amorphous  $Ni<sub>x</sub>Nb<sub>1-x</sub>$  alloys exhibit low-energy logarithmic behavior which is characteristic of electron scattering on the fast relaxing TLS's in full accordance with the Vladar-Zawadowski model.<sup>2</sup> In Ni<sub>59</sub>Nb<sub>41</sub> we found reproducible structures in the point contact spectra at high biases and higher ohmic contacts. We also studied the unusual features of superconducting  $Ni_{44}Nb_{56}$  contacts which can be explained by a percolation type of superconductivity, heating effects in the normal phase with increasing bias, and the influence of slow configurational changes close to the contact.

We acknowledge E. Sváb, A. Zawadowski, and I. K. Yanson for useful discussions. This work was supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO), the Stichting Fundamenteel Onderzoek der Materie (FOM), and by the Hungarian National Science Foundation under Grant No. T026327.

- <sup>1</sup> J. L. Black, in *Glassy Metals I*, edited by H. J. Guntherodt and H. Beck (Springer-Verlag, New-York, 1981), p. 167.
- $2$ K. Vladar and A. Zawadowski, Phys. Rev. B  $28$ , 1564 (1983);  $28$ , 1582 (1983); **28**, 1596 (1983).
- 3R. Harris and J. O. Strom-Olsen, in *Glassy Metals II*, edited by H. J. Guntherodt and H. Beck (Springer-Verlag, New-York, 1983).
- <sup>4</sup> I. K. Yanson and O. I. Shklyarevskii, Sov. J. Low Temp. Phys. 12, 509 (1986).
- 5D. C. Ralph and R. A. Buhrman, Phys. Rev. Lett. **69**, 2118  $(1992).$
- 6R. J. P. Keijsers, O. I. Shklyarevskii, and H. van Kempen, Phys. Rev. B 51, 5628 (1995).
- ${}^{7}R$ . J. P. Keijsers, O. I. Shklyarevskii, and H. van Kempen, Phys. Rev. Lett. **77**, 3411 (1996).
- 8O. P. Balkashin, R. J. P. Keijsers, H. van Kempen, Yu. A. Kolesnichenko, and O. I. Shklyarevskii, Phys. Rev. B **58**, 1294  $(1998).$
- <sup>9</sup>V. I. Kozub and I. O. Kulik, Zh. E<sup>ksp.</sup> Teor. Teor. Fiz. 91, 2243 (1986) [Sov. Phys. JETP 64, 1332 (1986)].
- 10O. Rapp, P. Lindqvist, and H. S. Chen, Solid State Commun. **54**, 899 (1985).
- 11C. J. Muller, J. M. van Ruitenbeek, and L. J. de Jongh, Physica C **191**, 485 (1992).
- <sup>12</sup>We did not reach low enough temperatures to investigate the relevance of the two-channel Kondo model, see J. von Delft, A. W. W. Ludwig, and V. Ambegaoker, Ann. Phys. (N.Y.) 273, 175 (1999).
- <sup>13</sup> S. Ami and K. Maki, Phys. Rev. B **19**, 1403 (1979).
- <sup>14</sup>W. L. Johnson, in *Glassy Metals I* (Ref. 1).
- <sup>15</sup>E. Sváb, S. Borbély, Gy. Mészáros, S. N. Ishmaev, and R. Glas, J. Phys. IV 3, 291 (1993).
- <sup>16</sup>A. Bardas and D. V. Averin, Phys. Rev. B **56**, R8518 (1997).
- 17Yu. G. Naidyuk, K. Gloss, and A. A. Menovsky, J. Phys. C **9**, 6279 (1997).