Reply to ‘‘Comment on ‘Weak-localization and Coulomb-interaction effects in hydrogen-doped Zr-Ni and Zr-Cu metallic glasses’’

Kokanović, Ivan; Leontić, Boran; Lukatela, Jagoda

Source / Izvornik: **Physical review B: Condensed matter and materials physics, 1992, 45, 5070 - 5071**

Journal article, Published version
Rad u časopisu, Objavljena verzija rada (izdavačev PDF)

Permanent link / Trajna poveznica: https://urn.nsk.hr/urn:nbn:hr:217:053012

Reply to "Comment on 'Weak-localization and Coulomb-interaction effects in hydrogen-doped Zr-Ni and Zr-Cu metallic glasses'"

I. Kokanović and B. Leontić
Department of Physics, Faculty of Science, University of Zagreb, P.O. Box 162, Zagreb, Republic of Croatia

J. Lukatela
Institute of Physics of the University, P.O. Box 304, Zagreb, Republic of Croatia
(Received 12 August 1991)

The alternative explanation for our magnetic-susceptibility results in terms of increased segregation of late transition elements due to the presence of hydrogen is ruled out because the hydrogen concentrations used are below the "critical" concentration for that process. Some other alternatives mentioned by Bakonyi were already discussed in our paper. But taking the magnetic-susceptibility results together with those of the temperature and magnetic-field dependence of electrical conductivity, we think that the most consistent picture was the one given in our original paper.

In his Comment on our paper [Phys. Rev. B **41**, 958 (1990)] Bakonyi is mainly concerned with those of our results pertaining to the magnetic susceptibility. He argues that the observed low-temperature upturn and its increase with hydrogen concentration can be explained in terms of localized moments of magnetic impurities and the increased segregation tendency of late transition elements due to the presence of hydrogen.

We think that the results and arguments which he quotes, although correct for the systems he mentioned, cannot be applied to our systems.

We take up Bakonyi's statements one by one.

(1) The statement that hydrogen can induce substantial atomic rearrangements which could lead to superparamagnetic Ni clusters in Zr-Ni glasses: The results he invokes [i.e., A. Cziraki *et al.*, J. Magn. Magn. Mater. **83**, 360 (1990)] refer to $Ni_{60}Zr_{40}$, while our system is $Ni_{33}Zr_{67}$, that is to say a Zr-rich system. It is well known (Ref. 16 in Bakonyi's paper and our Ref. 1) that the quantity of absorbed hydrogen is proportional to the Zr content because hydrogen atoms first occupy the tetrahedral sites coordinated with four zirconium atoms. Beyond a "critical" concentration hydrogen may cause a microphase separation. The very results of Cziraki *et al.* (mentioned above) confirm this because they have observed the formation of magnetic clusters for hydrogen-to-metal ratios greater than 0.97. It has to be noted that our maximum concentration in $Ni_{33}Zr_{67}$ is 0.33, and at such low concentration, we expect no clustering. This is confirmed by the results of Zehringer *et al.* (Ref. 2) whose x-ray-diffraction measurements on $Ni_{24}Zr_{76}$ with hydrogen concentration up to 0.46 show no changes which would point to a phase separation into Zr-hydride and a Ni-enriched alloy after hydrogen doping. The neutron diffraction experiments of Rodmacq *et al.* (Ref. 3) on $Cu_{50}Ti_{50}$ also showed no effect of clustering for hydrogen concentration up to 0.33, while the effect has been observed for a higher hydrogen concentration (0.84) (Ref. 16 in Bakonyi's paper).

Even if the effect of microclustering exists one would then expect a greater effect on the susceptibility in Zr-Ni system than in Zr-Cu system, because Cu atoms carry no magnetic moments, which is contrary to what we observe.

(2) As to the possible Fe impurities in the Zr-Cu system Bakonyi quotes Refs. 10 and 13 which show that sometimes 1 at. % Fe produces the effect of the low-temperature upturn and sometimes even 2 at. % does not, which he ascribes to different cooling rates during amorphization.

If such thermodynamic effects were present in our systems we would have observed similar behavior in the ZrCu and ZrNi systems, since they were produced in an identical way using the same Zr stock. It seems to us also that any impurity (Fe, Co) would have produced similar effects in ZrNi and ZrCu, but we observe that in fact these systems behave very differently. To explain the results as one "spurious" effect in one system and as another "spurious" effect in the other is really quite farfetched.

There is a more quantitative argument, namely, that from our magnetoresistivity data (Ref. 4) we can extrapolate H_Φ (a phase-coherent field) to $T=0$ K and from the intercept estimate the spin-scattering rate. This gives an upper limit of ≤ 1 ppm for possible magnetic impurities concentration.

(3) We agree, of course, that the behavior of χ_{expt} can be explained by invoking two diverse mechanisms using an equation such as Bakonyi's Eq. (1) in his Ref. 12, but this is not very helpful since the fitting parameter A reveals nothing pertaining to the microscopic origin of these mechanisms.

(4) As we pointed out in our paper, the Stoner factor for our systems is of the order $I \cong 0.35$, which is very far from 1. This is consistent with Batalla *et al.* (Ref. 5) who found $I=0.35$ and 0.15 for $Ni_{20}Zr_{80}$ and $Cu_{60}Ze_{40}$, respectively. They also found that λ_{SF} is negligible in Cu-Zr and small (<0.1) in Ni-Zr for concentrations of Ni less than 50 at. %. Obviously, if anything, the value of I

can only go down with hydrogen doping as has been explained in our paper (Ref. 4).

Concerning the value of I for ZrH_x, which Bakonyi assumes could be great, Papaconstantopoulos calculations (Ref. 6) show that it is $\cong 0.33$, so we cannot see how this could be a source of the scattering mechanism that Bakonyi suggests.

In conclusion, we may state that some of the alternative explanations offered may be viable. Indeed we have examined most of them ourselves and even mentioned some of them in our paper. But by taking all the results together, the most consistent and logical picture we could offer was the one given. Our research program will shortly expand to other systems and we hope the results will further clarify the behavior of these fascinating structures.

[1]K. Samwer and W. L. Johnson, Phys. Rev. B **28**, 2907 (1983); H. Kaneko, T. Kajitani, M. Hirabayashi, M. Ueno, and K. Suzuki, J. Less-Common Met. **89**, 237 (1983).

[2]R. Zehringer, E. K. Hlil, M. H. Tuilier, P. Oelhafen, and H. J. Güntherodt, J. Non-Cryst. Solids **117/118**, 425 (1990).

[3]B. Rodmacq, Ph. Mangui, and A. Chamberod, J. Phys. F **15**, 2259 (1985).

[4]I. Kokanović, B. Leontić, and J. Lukatela, Phys. Rev. B **41**, 958 (1990).

[5]E. Batalla, Z. Altounian, and J. O. Strom-Olsen, Phys. Rev. B **31**, 577 (1985).

[6]D. A. Papaconstantopoulos, Europhys. Lett. **15**, 621 (1991).