Comment on "Magnetic Correlations in Nanostructured Ferromagnets"

In a recent Letter [1], Löffler, Braun, and Wagner discuss the magnetic exchange coupling at interfaces in nanocrystalline (nc) ferromagnets based on small-angle neutron scattering (SANS) data for the grain-size dependence of a magnetic correlation length in nc Fe, Co, and Ni as a function of the grain size. We comment on the appropriateness of the data analysis and on the character of the interfaces in their samples.

In Ref. [1], the grain-size distribution is computed by analyzing the SANS data in terms of scattering by noninterfering hard spheres. It is known that interparticle interference becomes noticeable when the particle volume fraction x exceeds a few percent, and that methods which neglect the interference underestimate the grain size significantly, for instance by 30% at volume fractions as small as $x = 0.16$ [2]. It is now well established that grain boundaries in elemental nc metals are structurally similar to high-angle grain boundaries in macroscopic bicrystals, with the grain boundary "thickness" practically of atomic dimension [3]. Therefore, nc materials constitute *dense* and highly correlated arrangements of polyhedral grains with $x \approx 1$. While interparticle interference in moderately dense solutions ($x \approx 0.1$) affects only the scattering at small scattering vector q, computer simulations show that in dense nc solids the scattering at high q is also affected [4]. Since models based on noninterfering hard spheres ignore this effect, they are inadequate for determining grain-size distributions in nc solids.

Owing to the small scattering contrast of grain boundaries, the nuclear scattering in pore-free elemental nc ferromagnets is several orders of magnitude smaller than the magnetic scattering [5]. Since Ref. [1] reports a nuclear scattering cross section close in magnitude to the magnetic scattering, the former is most likely due to pores or oxides, not to grain boundaries. In fact, the authors of Ref. [1] previously reported significant porosity and oxidation for their materials [6]. Whereas Ref. [1] alludes to a general agreement between measurements of a mean grain size by SANS and by wide-angle x-ray diffraction, systematic studies by other authors found significant discrepancies between the two measures for the mean size, which supports our view that SANS measures the size of pores in inert gas condensed nanocrystalline materials, not the grain size [7]. Irrespective of this issue, standard wide-angle diffraction data analysis cannot validate the determination of the grain size *distributions,* because it provides no information beyond the mean size.

It is also pointed out that, based on SANS and magnetization data, the authors of Ref. [1] have concluded that their materials consist of nanoscale ferromagnetic clusters embedded in an interfacial phase of ferri- or antiferromagnetic oxides [6]. Therefore, although Ref. [1] is worded in terms of pure nanocrystalline metals, the conclusions regarding the magnetic coupling across interfaces pertain not to metallic grain boundaries, but to interfacial layers of ferri- or antiferromagnetic oxides.

Reference [1] reports a magnetic correlation length determined by analyzing the magnetic scattering signal in terms of scattering by noninterfering hard spheres. This presupposes that the magnetic microstructure is validly described by domains with uniform scattering contrast (uniform magnetization, **M**) and with discontinuous jumps of **M** at the domain boundaries. While the model of Ref. [1] allows for discontinuities in **M** at interfaces due to a nonuniform exchange constant, the experimental value of the effective exchange constant of nc Fe is reported identical to that of bulk Fe, which suggests an essentially uniform exchange interaction [compare discussion of Eq. (7) in [1]] and, therefore, a continuous variation of **M** at interfaces. Indeed, Eq. (8) of Ref. [1] predicts the characteristic length L of gradients in **M** to exceed 50 nm. In agreement with Herzer's theory [8] this implies wide domain boundaries and, consequently, a scattering law which is quite different [5] from that of the hard-sphere model. There is therefore good reason to suspect that the values of L derived from this model are erroneous.

Micromagnetics models have successfully predicted macroscopic magnetic properties of nanocrystalline materials [8], but it is of fundamental interest to verify their predictions on a microscopic scale. In this context the agreement between experiment and theory reported in Ref. [1] would be an important result. However, in view of the concerns about the data analysis, the result cannot be considered as conclusive.

J. Weissmüller[1] and A. Michels[2]
[1]Forschungszentrum Karlsruhe, 76021 Karlsruhe, Germany
[2]Universität des Saarlandes, 66041 Saarbrücken, Germany

Received 28 September 2000; published 18 September 2001
DOI: 10.1103/PhysRevLett.87.149701
PACS numbers: 75.50.Kj, 61.12.Ex, 75.10.Nr, 75.60.Ch

[1] J. F. Löffler, H. B. Braun, and W. Wagner, Phys. Rev. Lett. **85**, 1990 (2000).
[2] W. S. Rothwell, J. Appl. Phys. **39**, 1840 (1968); G. Kostorz, in *Small Angle X-ray Scattering,* edited by O. Glatter and O. Kratky (Academic Press, London, 1982), p. 471.
[3] E. A. Stern *et al.,* Phys. Rev. Lett. **75**, 3874 (1995); F. Boscherini, S. de Panfilis, and J. Weissmüller, Phys. Rev. B **75**, 3365 (1998).
[4] I. Detemple *et al.,* Scr. Mater. **37**, 1685 (1997).
[5] J. Weissmüller *et al.,* Mater. Res. Soc. Symp. Proc. **457**, 231 (1997); A. Michels *et al.,* J. Appl. Phys. **87**, 5953 (2000).
[6] J. F. Löffler *et al.,* Phys. Rev. B **57**, 2915 (1998); J. Löffler *et al.,* Physica (Amsterdam) **241B–243B**, 603 (1997).
[7] P. G. Sanders, J. A. Eastman, and J. R. Weertman, Acta. Mater. **46**, 4195 (1998).
[8] G. Herzer, IEEE Trans. Magn. **26**, 1397 (1990).

