ever, we noticed that there are three luminescence peaks with exceptionally strong temperature dependence, the cause of which is yet to be identified.

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Direct Observation of an Amorphous Spin-Polarization Distribution*

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Evidence is presented from neutron diffraction measurements for the existence of an amorphous spin-polarization distribution in a sputtered rare-earth–iron alloy of composition 33 at.% Tb, 67 at.% Fe.

We have made neutron diffraction measurements on a ferromagnetic sputtered sample of composition 33 at.% Tb, 67 at.% Fe, which allow the first direct observation of the spin polarization in an amorphous magnetic material.

Bulk magnetization measurements[1] show that the material is actually ferromagnetic, in the sense that a macroscopic spontaneous moment is observed. The magnetic isotherms shown in Fig. 1(a) indicate a high degree of saturation in fields of 2–3 kOe, followed by a high-field susceptibility. The zero-field intercepts are plotted versus temperature in Fig. 1(b), indicating a well-defined Curie point of 388 K. This value, as well as the spontaneous moment $\sigma(0, 0) = 2.8\mu_B/$mole, are substantially lower than that of the crystalline $TbFe_2$ Laves phase (710 K and $4.7\mu_B$, respectively).[2] Both these features were also found in ternary Fe-Pd-P alloys[3] that are believed to be amorphous. The decrease in $\sigma(0, T)$ below 100 K in Fig. 1(b) is anomalous but may partly result from the procedure used in subtracting the susceptibility term. Prolonged annealing of the sample in the neighborhood of 650 K produces an increase in magnetization with time, providing evidence for the partial recrystallization of the $TbFe_2$ Laves phase at this temperature.

The neutron data were taken at the National Bureau of Standards Reactor on a disk 25 mm diam by 1 mm thick with 1.36-Å incident wavelength, the collimation being 20′ in-pile, 0 between monochromator and sample, and 40′ before the counter. Diffraction peak widths were unaltered by inserting 20′ collimation before the counter, and therefore instrumental resolution corrections were neglected. Diffraction patterns taken at 423 and 4 K, with instrumental

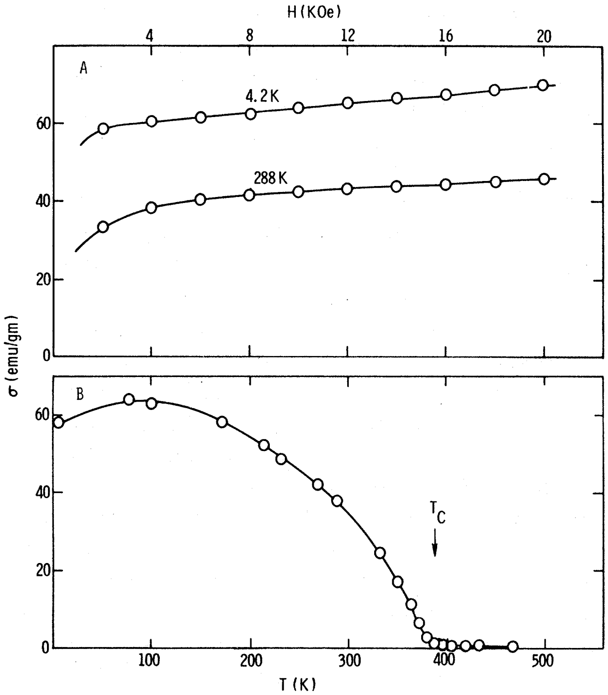

FIG. 1. (a) The magnetic moment of sputtered $TbFe_2$ versus field at two temperatures. (b) The spontaneous magnetization versus temperature. The measurements from room temperature up were made in a constant 1-kOe field and scaled to the extrapolated data at 290 K.

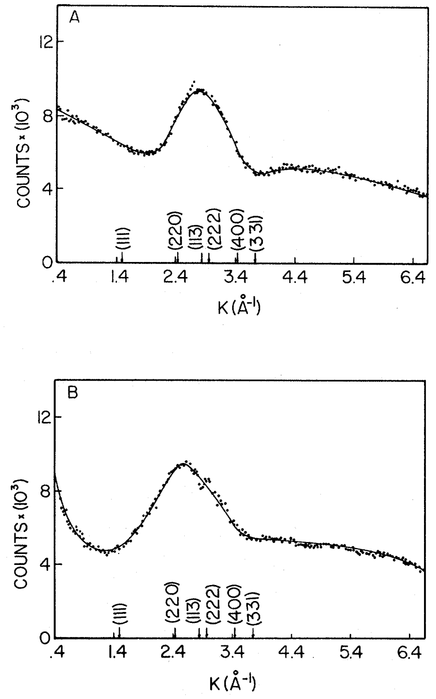

FIG. 2. Neutron diffraction patterns of sputtered $TbFe_2$ (a) at 423 K [the small pip near the top of the broad peak is the remainder of an Al (111) peak due to the radiation shield]; (b) at 4.2 K.

background subtracted, are shown in Fig. 2. The arrows indicate the positions of the Bragg lines in the corresponding Laves-phase compound.

The high-temperature pattern, Fig. 2(a), is consistent with scattering of atomic origin from an amorphous structure, superimposed on a form-factor–like background due to paramagnetic diffuse scattering. The broad maximum is very near the angular position of the (113) and (222) reflections of $TbFe_2$, the strongest nuclear peaks of the crystalline phase. The broadness of the peak ($\sim 13°$ in 2θ) cannot be attributed to microcrystallinity, however, for several reasons. The pattern is not characteristic of a Laves phase with broadened lines, since there would also be peaks at much lower and higher angles than the observed maximum. If we are dealing with a different phase altogether, application of the Scherrer formula would indicate a crystallite size of 6–8 Å, i.e., comparable to several interatomic spacings. Not only is crystallinity on such a scale operationally indistinguishable from the amorphous state, but it would give rise to a comparably broad small-angle scattering, which we do not observe. As far as the atomic structure is concerned, we therefore conclude that the sample is indeed amorphous and reasonably homogeneous, and that any microcrystalline phases, if present, are below the level of observation.

Turning to the low-temperature data in Fig. 2(b), we note that the diffuse peak appears to have broadened and shifted to a significantly lower angle; in addition, a small-angle component to the scattering has arisen. Both these changes are attributable to the development of the macroscopic ferromagnetic polarization. In order to make these features more visible, we subtracted the high-temperature data from the low, obtaining the difference pattern reproduced in Fig. 3.

This pattern, if we assume that no strong magnetic correlations remain above T_c, reflects the spatial distribution of the spin polarization alone. It is in all respects characteristic of an amorphous structure, with the exception of the small-angle "tail." (It is to be noted that this subtraction procedure automatically gives the modified intensity[4] needed to obtain the radial distribution function of the moment centers by Fourier transformation, since the total scattering remains as a paramagnetic contribution at high temperature;

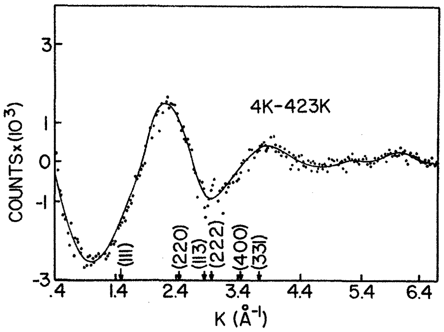

FIG. 3. Difference pattern (low-temperature data minus high) showing the purely magnetic intensity for sputtered $TbFe_2$.

moreover, corrections such as absorption, nuclear incoherent scattering, and inelastic scattering, to the extent that it is temperature independent, are canceled out.) The data in Fig. 3 therefore offer convincing proof for the existence of a truly amorphous spin-polarization distribution in this alloy. Again, any microcrystalline magnetic phases are below the level of observation.

Inasmuch as the composition is binary, an unambiguous interpretation of the data by Fourier methods is in principle impossible, since they result from a superposition of three pair distribution functions in both space and spin coordinates. Even if the fact that the atomic distribution peaks near the strongest peak of $TbFe_2$ is used as a starting point for a model of the atomic correlations, the unknown magnetic moment and degree of localization of the randomly distributed rare-earth and iron moments would render direct methods of interpreting the spin polarization problematic.

Nevertheless, several simple models of the spin distribution would appear to be inconsistent with our data. The fact that the first maximum of the spin distribution is shifted from that of the atomic distribution rules out the admittedly simple ferromagnetic model in which each atomic site carries the same average magnetic moment. Similarly, if the immediate neighborhood of any site looked like the ferrimagnetic arrangement in the crystalline $TbFe_2$ phase (rare-earth and Fe moments antiparallel), a maximum near the (111) position, the strongest magnetic reflection in the crystalline material, might be expected. More realistic models of the spin distribution cannot be constructed without a detailed knowledge of the atomic correlations.

The small-angle "tail" observed in Fig. 2(b), which is absent in 2(a), arises from the broadening of the (000) magnetic peak. This indicates that some sort of inhomogeneity of a longer range than the average near-neighbor spin correlations is present in the magnetic structure, which cannot be related to any inhomogeneity in the atomic structure. This small-angle scattering is unusually broad and hence must be interpreted on the basis of a small-scale "graininess" or clustering in the magnetic structure rather than domains in the conventional sense. These phenomena are being studied further as functions of temperature and magnetic field to see whether they are characteristic of a sputtered sample or are perhaps intrinsic to the magnetic amorphous state. It will obviously be of great interest also to determine whether critical scattering at the phase transition or localized spin-wave excitations can occur in such materials.

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