## Detection of a magnetic viscosity in a spin glass through small-angle neutron scattering in a magnetic field

B. N. Mokhov

Institute of Precision Alloys, I. P. Bardin Central Scientific-Research Institute of Ferrous Metallurgy

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A slow relaxation of spatial magnetic inhomogeneities, which gives the system a magnetic viscosity, has been observed for the first time in a spin glass. The experimental method was to study the time dependence of the small-angle magnetic scattering of neutrons in the spin glass  $Fe_{65}Ni_{25}Cr_{10}$  during the application and removal of a magnetic field. A possible mechanism for the magnetic viscosity is discussed with reference to a possible double transition paramagnet  $\rightarrow$  ferromagnet  $\rightarrow$  spin glass.

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A spin-glass state occurs in 3d alloys near a concentration ferromagnetic—antiferromagnetic transition.<sup>1</sup> The magnetic phase diagrams of such systems<sup>2,3</sup> are like those of classical spin glasses<sup>4</sup> in that the magnetic state has a twofold nature at concentrations just beyond the ferromagnetic threshold: As the temperature is lowered, there is initially a transition from a paramagnetic state to a ferromagnetic state (F), and then a spin glass (SG) appears. It has not yet been resolved, however, whether the spin glass forms as a result of the  $F \rightarrow$  SG transition as the temperature is changed<sup>5,6</sup> or whether it forms along with the ferromagnetic state.<sup>7,8</sup> We have carried out experiments to determine whether the use of small-angle neutron scattering in a magnetic field to study a characteristic property of a spin glass—its magnetic viscosity—would yield direct evidence on the dynamics of the spatial magnetic structure and thus on the mechanism for the formation of the spin glass.

We studied the  $\gamma$ -phase alloy 65 at.% Fe, 25 at.% Ni, 10 at.% Cr from a quasibinary cut of Fe<sub>65</sub> (Ni<sub>1-x</sub>Cr<sub>x</sub>)<sub>35</sub> (Refs. 3 and 9). The diffraction patterns were recorded on a neutron diffractometer with a wavelength of 1.59 Å in a helium cryostat. A superconducting solenoid could produce a magnetic field in the direction perpendicular to the scattering vector.

This particular composition was chosen because it allows the  $F \rightarrow SG$  transition as the temperature is changed (Fig. 1a), while it also maximizes the small-angle neutron scattering<sup>3</sup> and the magnetic viscosity.<sup>10</sup> It is important to note that this composition is similar to that of an alloy in which the low-temperature magnetization anomaly which reflects the  $F_{65}$  Ni<sub>25</sub> Cr<sub>10</sub> transition is suppressed by a magnetic field of about 2 kOe (Refs. 11 and 12).

Figure 1b shows the results of the measurements, which were carried out in the following order: 1—The sample was cooled to 4.2 K in the absence of an external

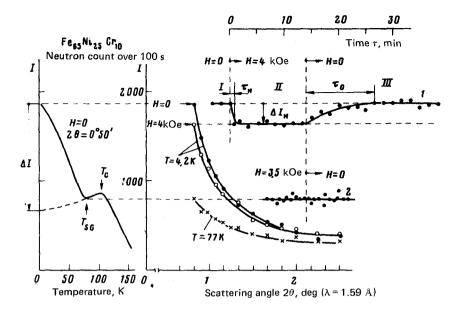


FIG. 1. Small-angle neutron scattering in the spin glass  $Fe_{65}Ni_{25}Cr_{10}$ . a—Temperature dependence of the intensity with H=0 and  $2\theta=0^{\circ}50'$  (Ref. 3); b—angular dependence at 4.2 K.

•) H=0; •) H=4 kOe; ×) 77 K. Curves 1 and 2 were recorded at  $2\theta=0^{\circ}50'$  during the imposition and removal of the magnetic field at 4.2 and 77 K, respectively.

magnetic field. 2—The angular dependence of the neutron scattering was measured at 4.2 K and H=0. 3—The neutron counter was placed at the angle  $2\theta=0^{\circ}50'$ ; after several control points were measured at H=0 (see part I of curve 1 in Fig. 1b), a magnetic field of 4 kOe was applied, and the time dependence of the intensity was measured (region II). 4—The angular dependence of the neutron scattering was measured at 4.2 K and H=4 kOe. 5—The counter was returned to its original position,  $2\theta=0^{\circ}50'$ ; after the field was removed, the time dependence of the intensity was measured with H=0 (region III).

The entire measurement cycle was carried out three times to eliminate systematic errors.

If the basic state of this alloy were a spin glass formed in an  $F \to SG$  transition, we would expect the intensity of the small-angle scattering at 4.2 K to be reduced by a factor of 2-2.5 ( $\Delta I$  in Fig. 1a) by a magnetic field of 4 kOe (this is twice the field which suppresses the  $F \to SG$  transition<sup>11,12</sup>). We see from Fig. 1b, however, that the actual intensity decrease  $\Delta I_H$  is only about 10%. It follows that the magnetic field, which was found to suppress the  $F \to SG$  transition, causes only very slight change in the degree of magnetic inhomogeneity and the spatial structure of the spin glass. We interpret these results as evidence against a double  $F \to SG$  transition.

Curve 1 in Fig. 1b shows the relaxation to the equilibrium spatial structure at 4.2 K. The viscous change in the magnetic structure, as in the magnetization, <sup>10</sup> is not seen in strong magnetic fields. The relaxation time after the field is turned on is  $\tau_H < 40$  s, while that after the field is turned off is  $\tau_0 \sim 10$  min. We might note that

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in measurements<sup>10</sup> of the magnetic viscosity of samples of similar composition the magnetization continued to vary for 30 min or more.

After the sample was heated in a zero field to 77 K, corresponding measurements were carried out in a field of 3.5 kOe. In this case, however (curve 2 in Fig. 1b), the magnetic field does not affect the small-angle scattering. This result is in agreement with the data of Ref. 10, where measurements at 77 K also failed to reveal a magnetic viscosity. These facts can be explained on the basis of a solidification temperature  $T_{\rm SG} = 75$  K for this spin glass (Fig. 1a).

Analysis of all of these results suggests that the magnetic field causes only a partial transition in this system, to a spatially more homogeneous ferromagnetically ordered state.

The magnetic viscosity detected in these neutron-diffraction measurements after the removal of the magnetic field was caused by a slow, reversible  $F \to SG$  transition. The small values of  $\Delta I_H$  and  $\tau_0$ , however, show that the primary mechanism for the viscosity observed in the magnetization measurements of Ref. 10 was a slow relaxation of the magnetization of a subsystem of a cluster spin glass proper.

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