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Transverse magnetism of the diluted antiferromagnet $Fe_{1-x}Mg_xBr_2$ ($x \sim 0.15$)

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Abstract

Neutron diffraction and magnetization measurements of the diluted crystal $Fe_{1-x}Mg_xBr_2$ ($x \sim 0.15$) were performed. Successive phase transitions at zero applied field at $T_{N1} \sim 12$ K and $T_{N2} \sim 10$ K were found. A transverse ferromagnetic moment was detected just below T_{N2}. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Neutron diffraction; Magnetization; Transverse magnetism; Ising antiferromagnet

Typical metamagnetic characteristics are known to appear in layered compounds with strong Ising anisotropy and ferromagnetic intralayer and antiferromagnetic interlayer-interactions, such as $FeCl_2$. Iron bromide $FeBr_2$ was also believed to be a metamagnet. It undergoes a field-induced metamagnetic phase transition at lowest temperatures and shows noncritical fluctuations around the multicritical point. These properties originate from the Ising character of the spins. However, recent studies of the magnetization [1] and the neutron diffraction [2] with tilted samples revealed that the transverse spin component also contributes to the phase transitions. The drastic change of transverse components can explain the sharp anomaly observed in the specific heat [3].

In order to perform a systematic study for the mechanism of phase transitions in $FeBr_2$, the diluted crystals $Fe_{1-x}Mg_xBr_2$ with effectively weak Ising anisotropy and weak interactions were prepared. Karszewski et al. had already studied the diluted sample with $x \sim 0.15$ by the Faraday rotation technique with axial fields and reported the existence of a field-induced spin-flop phase with a finite range of inner field, instead of the metamagnetic coexistence [4]. Recently, a sample with different concentration $x \sim 0.05$ was investigated by Petracic using SQUID magnetometry and specific heat [5,6]. He proposed a novel field-temperature phase diagram which contains a spin-flop phase for only finite temperatures (above ~ 3 K) and the metamagnetic phase transition at lowest temperatures (below ~ 3 K). In these studies of both diluted samples, the possible direct detection of the transverse moment was, however, not performed. In the present study, the transverse moment of the $x \sim 0.15$ sample was investigated using neutron diffraction and SQUID measurements.

The neutron diffraction was performed using the D15 spectrometer installed at ILL in Grenoble with a vertical magnet for applying the field parallel to the *c*-axis. The *ab*-plane of the sample was horizontally placed by centering of the measured ten Bragg peaks with indices $(1,1,0)$, $(2,2,0)$ and their equivalent points. The mechanism of the detector-cradle of this spectrometer enabled to measure the off-plane Bragg peaks at $(h,k,1/2)$. The intensity of all peaks was measured by taking omega-scans. Typical widths of the peaks were 0.5° for $(h,k,0)$ and 1° for $(h,k,1/2)$.

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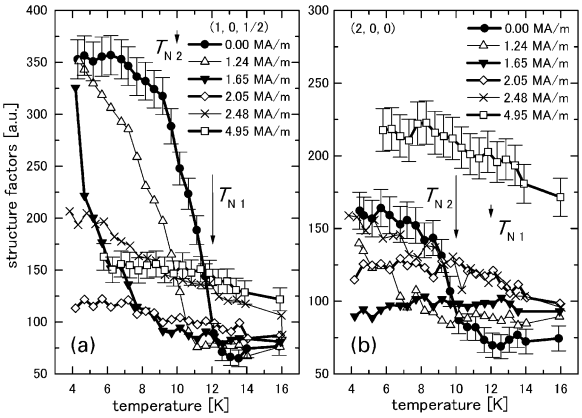

Fig. 1. Temperature dependence of the structure factors at zero and applied fields for (1, 0, 1/2) (a) and (2, 0, 0) (b). The transition temperatures at $H = 0$ were indicated as $T_{N1}(\sim 12\,K)$ and $T_{N2}(\sim 10\,K)$. For $\mu_0 H$ in T unit, $\mu_0 = 1.2566\,T(MA/m)^{-1}$ should be multiplied.

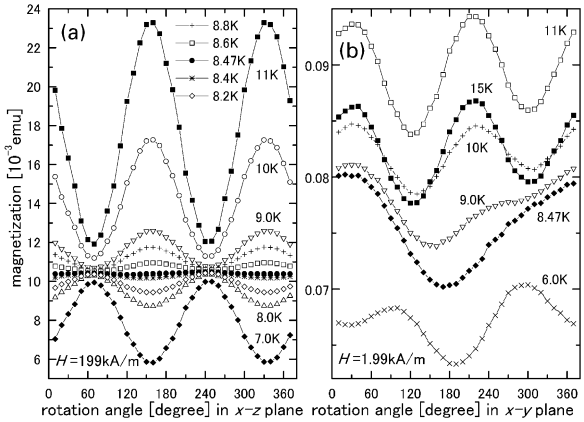

Fig. 2. Angular dependence of the magnetization of the sample rotated in the x–z plane at (a) $H =$ 199 kA/m ($\mu_0 H = 0.25\,T$) and (b) in the x–y plane at $H = 1.99$ kA/m ($\mu_0 H = 2.5 \times 10^{-3}\,T$).

As shown in Fig. 1, successive phase transitions under zero field were found at $T_{N1} \sim 12\,K$ and $T_{N2} \sim 10\,K$. By applying low fields $H < 2.0$ MA/m (~ 25 kOe), both critical temperatures were decreased, as shown in Fig. 1. At $T = 6\,K$, the intensity of the (1, 0, 1/2) peak showed a minimum at $H \sim 2.0$ MA/m, which can correspond to the spin-flop transition [4]. However, the transition to the saturated paramagnetic phase was smeared even in the plots of the field dependence because of an unclear contribution to the neutron intensity at higher fields ($H > 2$ MA/m). This contribution does not come from any magnetic ordering: the reasons are as follows. (a) It does not decrease above T_{N1}. (b) At some $(h, k, 1/2)$ points, the unexpected 'background' peak (a peak at zero field and above T_{N1}) was observed at an off-centered position in the omega-scan, and at the same position the peak at high fields appeared.

It is obvious that antiferromagnetic ordering of the longitudinal component occurs at T_{N1}. The lower-temperature transition at T_{N2}, which was not observed in the previous study [4] might be due to the transverse ferromagnetic ordering, because an order-parameter-like temperature dependence was found at the nuclear Bragg position, and the applied axial fields decreased the intensity.

In order to find the transverse ferromagnetic component directly in magnetometry, SQUID measurements were performed. A quantum design MPMS5 system using the DC-SQUID detection method with a horizontal-rotation sample holder was employed. Here the measured magnetization was always the component parallel to the vertical applied field. A measurement of the rotation angle dependence of the background magnetization from the sample holder at corresponding temperatures and fields proved to yield negligible contributions in comparison with the sample magnetization. A sample crystal with a size of $\sim 1.5 \times 1.5 \times 0.2\,mm^3$ was mounted using Apiezon N grease.

Firstly, the angular dependence of the magnetization under constant applied fields $H = 199$ kA/m (2500 Oe) and 1.99 kA/m (25 Oe) and fixed temperatures was measured for the sample rotation along the x–z and y–z planes, where the x-, y- and z-axis indicate the three orthogonal directions of the sample crystal and the z-axis was the hexagonal c-axis. Fig. 2(a) shows representative curves. The data around T_{N1} or above showed a maximum at $H \| z$ ($\chi_\| H$) and a minimum at $H \perp z$ ($\chi_\perp H$). Since $\chi_\perp$ is almost constant, but $\chi_\|$ drastically decreases at decreasing temperatures, the amplitude of the component of 180° periodicity decreased with decreasing temperature and changed the sign. The measurement at 8.47 K showed the smallest amplitude of the 180° component.

Secondly, the same sample was remounted to measure the angular dependence in the x–y plane, which is shown in Fig. 2(b). Here the angle-independent part is due to the temperature dependence of $\chi_\perp$. Due to the small misalignment of the sample mounting, a 180° periodicity was observed at higher temperatures (10, 11 and 15 K). This component was small at 9 K, negligible at 8.47 K and obtained a negative sign at 6 K. Then, the transverse ferromagnetic moment which must be small and can appear only below T_{N2} was expected to be detectable at those temperatures. In addition to the background component of the 360° periodicity at higher temperatures, another 360° part with shifted phase was observed below T_{N2} which showed some temperature dependence. It is plausible that the latter contribution comes from the transverse moment.

To the best of our knowledge, the strong increase of the intensity of the neutron diffraction induced by high fields is an unknown phenomenon. Its origin seems to be related to the background peaks at $(h, k, 1/2)$, which possibly refer to a kind of lattice distortion. Additional neutron-scattering experiments will be necessary for a thorough understanding.

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