Neutron scattering study of transverse magnetism in the metamagnet FeBr₂

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Abstract. In order to clarify the nature of the additional phase transition at \(H_1(T) < H_c(T)\) of the layered antiferromagnetic (AF) insulator FeBr₂ as found by Aruga Katori et al. (1996) we measured the intensity of different Bragg-peaks in different scattering geometries. Transverse AF ordering is observed in both AF phases, AF I and AF II. Its order parameter exhibits a peak at \(T_1 = T(H_1)\) in temperature scans and does not vanish in zero field. Possible origins of the step-like increase of the transverse ferromagnetic ordering induced by a weak in-plane field component when entering AF I below \(T_1\) are discussed.

PACS. 75.25.+z Spin arrangements in magnetically ordered materials (including neutron and spin-polarized electron studies, synchrotron-source X-ray scattering, etc.) – 75.30.Kz Magnetic phase boundaries (including magnetic transitions, metamagnetism, etc.) – 75.50.Ee Antiferromagnetics

1 Introduction

Metamagnets with large uniaxial anisotropy like FeCl₂ and FeBr₂ are widely accepted as model systems for investigating fundamental properties of antiferromagnets. Both compounds are ionic insulators with localized magnetic moments at the Fe\(^{2+}\)-sites, arranged in triangular layers. The inset of Figure 1 shows the hexagonal unit cell of FeBr₂ (space group \(D_{3d}^1 = P3m1\), Néel temperature \(T_N = 14.1\) K), where adjacent (001) layers of Fe\(^{2+}\) ions are separated by two layers of Br⁻ ions. The spin directions at low temperatures, \(T \ll T_N\), and in zero external magnetic field, \(H\), are conventionally assumed to point parallel and antiparallel to [001], respectively, from layer to layer. Thus a Néel type ground state with “up” and “down” spin sublattices seems to emerge as in the case of FeCl₂ (space group \(D_{3d}^5 = R3m\), \(T_N = 23.7\) K). However, while FeCl₂ – when exposed to an axial magnetic field \(H\) – reveals a classic tricritical point on its \(H-T\) phase line [1,2], FeBr₂ behaves in a more complicated fashion (Fig. 1).

Similarly as in FeCl₂ the lines \(H_{c1}\) and \(H_{c2}\) denote the phase transition of first-order from AF long-range order to the paramagnetic (PM) saturated phase via a mixed phase (AF + PM). However, above the multicritical point (MCP) temperature, \(T_{MCP} = 4.6\) K, apart from the critical phase line, \(H_c(T)\), regions of strong non-critical fluctuations are encountered. They are peaking along novel lines denoted as \(H_{\perp}(T)\) and \(H_{\parallel}(T)\), respectively [3]. Between \(H_{\perp}\) and \(H_{\parallel}\) the hyperfine field acting on the down-spin sublattice gradually varies from extreme negative to positive values, thus indicating strong transverse spin precession on the Mössbauer spectroscopic time scale [4]. In addition, another line, \(H_1(T)\), is observed in the vicinity of \(H_{\perp}(T)\). It was discovered by Aruga Katori et al. [5] in specific heat data and depicts a hitherto unexpected first-order phase transition. In magnetization measurements, using magnetic fields tilted with respect to the layer normal, it manifests itself as a slightly shifted line, \(H_1'(T)\) [6].

Recently, attention has been focused on the investigation of the nature of the phase transition at \(H_1(T)\). It divides the AF phase region into two sub-phases AF I \((H < H_1)\) and AF II \((H_1 < H < H_c)\). From magnetization measurements in tilted fields [6] it was conjectured that transverse spin ordering exists in the AF II phase, with a jump of the axial magnetization component \(M_{ax} = M_z\) at \(H_1'(T)\), provided that the transverse \((i.e.\ in-plane)\) magnetization, \(M_{pl}\), is aligned by an auxiliary field, \(H_{pl}\). In order

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to explain the obvious coupling between the in- and out-of-plane spin polarization an off-diagonal exchange interaction between planar and axial spin components was postulated, which is allowed in triclinic point symmetry [7].

Within the framework of a simple Néel state model with rigid spins the discontinuous enhancement of the ferromagnetic (FM) order parameter should give rise to a reverse jump in the AF order parameter. However, this was not clearly confirmed by elastic neutron scattering measurements. Apart from criticality at the phase boundary, \( H_{c}(T) \), and non-critical fluctuations at \( H_{c} \) and \( H_{c} \), only a weak anomaly of the AF order parameter is evidenced at \( H_{c}(T) \) by virtue of the temperature and magnetic field dependencies of the \((2, 0, 1/2)\) neutron scattering intensities when scaled with \( T_{c}(H) \) or \( T_{1}(H) \) [8]. Thus, in order to solve the enigmatic situation and to clarify the characteristics of \( H_{c}(T) \) it appears useful to investigate more carefully the spin structure, viz. in particular the planar components of the AF order parameter. To this end novel experiments of neutron scattering with magnetic fields both parallel and tilted to the \( c \) axis have been done. For the first time they evidence that the ground state spin configuration of FeBr\(_{2}\) in zero external field exhibits a transverse component of the AF order parameter in all AF phases, \( i.e. \) at \( H < H_{c1} \). As a signature of the phase transition at \( H_{c1}(T) \) it shows a peak together with a step-like rise of the transverse FM moment in the tilted configuration.

2 Experimental details

Neutron scattering measurements were performed at the High Flux Reactor at the Institut Laue Langevin (ILL) in Grenoble, France, and at the Berlin Neutron Scattering Center of the Hahn-Meitner Institut (HMI) in Berlin, Germany.

In the ILL experiments we used the D15-instrument as a two-axis thermal neutron normal-beam diffractometer with a cryostat containing a vertical superconducting magnet (\( \mu_{0}H \leq 6\) T). The neutron wavelength was \( \lambda = 1.176\) Å with a \( \lambda / 2 \) contamination of about 0.1%. It should be remarked that a non-horizontal scattering plane was chosen in some experiments. In those cases we changed the detector tilt angle, \( \nu \), for the different Bragg peaks. Almost all data shown in this paper are obtained from omega scans. We used two FeBr\(_{2}\) samples with sizes \( 20 \times 15 \times 10 \) mm\(^3\) and \( 4 \times 6 \times 5 \) mm\(^3\), mounted either with the \( c \) axis parallel to the applied field or tilted under an angle \( \theta \approx 30^\circ \) with respect to the field axis. Magnetization measurements show that for angles \( \theta < 30^\circ \) the tilting has only a small influence on the phase transition at \( H_{c}(T) \). It seems that only the component \( H \cos(\theta) \) drives the global critical behavior [6].

At HMI we used the E4-instrument employing a sample with size \( 6 \times 6 \times 2 \) mm\(^3\). It was mounted with its \( c \) axis parallel to the applied field, \( \mu_{0}H \leq 6\) T, which was supplied by a horizontal superconducting magnet.

In this configuration it was straightforward to measure the \((0, 0, 1/2)\) Bragg reflection, where both the \( c \) and the field axis lay in the scattering plane. The neutron wavelength was \( \lambda = 2.4\) Å with the collimator condition “40̊-40̊-open-40̊”.

Magnetization and ac susceptibility data were obtained using a SQUID magnetometer (Quantum Design MPMS5S).

3 Experimental results

3.1 Scattering data from ILL

Figure 2 shows the measurements of the scattering intensity \( I \) vs. temperature \( T \) for the \((2, 0, 1/2)\) Bragg peak, which is a measure of the axial AF order parameter. The \( c \) axis is parallel to the applied field, \( H \), which is varied from \( H = 0 \) (curve 1) to \( H = 2.39\) MA/m (curve 5).

Two features are obvious: In agreement with previous work [8] the phase transitions at \( T_{c}(H) \) are clearly indicated by points of inflexion in \( I(T) \) (arrows) in accordance with those found in the magnetization curves, \( M(T) \), two of which are shown in Figure 2 for \( H = 1.91\) MA/m (curve 3’) and 2.39 MA/m (curve 5’). Furthermore the anomalous decrease of the intensity due to non-critical fluctuations is observed at \( H_{c}(T) \), where the spin-down sublattice switches into an almost non-magnetized state, while the spin-up sublattice remains highly magnetized along the applied field. This causes an anomalous increase of \( M \) vs. \( T \) and a reduction of the AF order parameter.

![Figure 1. H-T (B-T) phase diagram of FeBr\(_{2}\) presented by interpolated lines and data points [3,6, where H (B = \( \mu_{0}H \)) is the applied axial magnetic field (induction). H\(_{c1}\) is the second-order phase line, while H\(_{c2}\) and H\(_{c3}\) are the boundaries of the mixed phase AF + PM. Dashed lines are extrapolated, which is allowed in trigonal point symmetry [7].](image)