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Unusual field-induced transition in a frustrated itinerant antiferromagnet

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A model that includes instability of the magnetic moment in a frustrated lattice has recently been proposed for RMn_2 systems (R = rare earth). An ordered mixed phase induced by a magnetic field in which magnetic and nonmagnetic sites coexist has been predicted by Monte Carlo simulations. The existence of this transition is now confirmed by a neutron-diffraction study of $TbMn_2$ in a magnetic field.

The interesting physics provided by frustration in antiferromagnetic systems is under active discussion in the literature. Frustration has been widely studied in disordered systems (spin glasses). It can also occur in both metallic and insulating periodic systems, either because it is inherent to the crystal structure, or due to competing interactions. The low-temperature behavior of such systems is very rich because of the existence of many states with similar energies. In rare-earth (R) metals helimagnetic (Tb, Dy) and antiphase (Tm) ordering have been recognized for a long time. More recently, other unusual behaviors have been reported: $Gd_3Ga_5O_{12}$ remains disordered even at low temperature, "order from disorder" has been proposed for $Fe_2Ca_3(GeO_4)_3$ and for the kagomé lattice $SrCr_8Ga_4O_{19}$.[1]

In all the previous cases all relevant sites are magnetic. In recent work,[2] however, in order to explain the peculiar properties of RMn_2 systems another possibility has been considered: the instability of the magnetic moment. Frustration near the magnetic-nonmagnetic (M-NM) transition yields complex magnetic-ordered phases in which magnetic and nonmagnetic sites coexist and which show unusual dependence on external parameters such as magnetic field, temperature, applied pressure or alloying. A similar situation can occur in frustrated Ce compounds due to the Kondo effect or in compounds where the lowest crystal-field level is a singlet.

Since in the RMn_2 compounds the R-R and R-Mn exchange interactions are an order of magnitude smaller than the Mn-Mn interactions, we restrict the discussion to the Mn lattice. Magnetic properties of the Mn lattice are then described by a Hubbard model, where the on-site Coulomb repulsion U is of the order of the bandwidth W, close to the M-NM instability. As the anisotropy of the Mn sites is very large transverse fluctuations of the Mn moment are neglected and only the longitudinal component of the local moment, $\mu_i = \langle n_{i\uparrow} - n_{i\downarrow} \rangle$, is considered. The effective Hamiltonian can be mapped into a Blume-Capel Hamiltonian:[2]

$$H = \Delta \sum_i S_i^2 + 1/2 \sum_{i \neq j} J_{ij} S_i S_j$$

where Δ and J_{ij} are related to the parameters of the Hubbard model. Close to the M-NM transition $\Delta > 0$ and S_i can take three values: $S_i = \pm 1$ if the site has a magnetic moment $\mu_i = \pm \mu$, and $S_i = 0$ if $\mu_i = 0$. We take antiferromagnetic nearest-neighbor interactions $J_1 > 0$, the second-neighbor interactions J_2 can be > 0 or < 0. Rather than introduce the complexity of the Laves phase RMn_2 structure, a triangular lattice has been used to model frustration. This model leads to a rich phase diagram that can be related to different RMn_2 compounds: (a) Nonmagnetic phase where $\mu_i = 0$ in all Mn sites ($ScMn_2$, $ErMn_2$); (b) magnetic phases in which $\mu_i \neq 0$ in all Mn sites (YMn_2, $NdMn_2$); and (c) a new mixed magnetic phase ($ThMn_2$, $DyMn_2$) in which because of frustration the molecular field on some Mn sites, but not all, is too small to compensate the energy Δ necessary to stabilize the moment. The effect of an applied magnetic field is interesting: Several transitions are induced, their number depending on the initial state in zero field. Furthermore, Monte Carlo simulations show an unexpected feature: At some critical fields the number of magnetic sites decreases with increasing field. Figure 1 shows the evolution of an

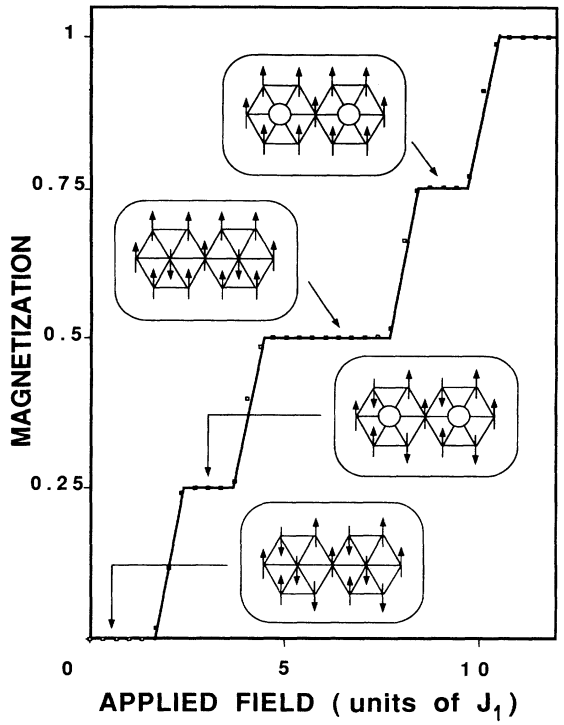

FIG. 1. Monte Carlo simulation for increasing magnetic field, for $\Delta=J_1$ and $J_2=J_1/2$, corresponding to an antiferromagnetic phase in zero field.

antiferromagnetic initial state: four transitions are observed at $h_1=-\Delta+2(J_1+J_2)$, $h_2=\Delta+2(J_1+J_2)$, $h_3=-\Delta+6(J_1+J_2)$, and $h_4=\Delta+6(J_1+J_2)$. Above the first and the third critical fields mixed phases are obtained.

We now report on the observation of this unusual type of transition in neutron-diffraction experiments on $TbMn_2$ in a magnetic field. The experiments were carried out on a single crystal of $TbMn_2$ in fields up to 6.5 T on the $D15$ diffractometer at Institut Laue-Langevin (ILL), Grenoble. They confirm the prediction of a field-induced mixed phase. Full details of this study will be published elsewhere,[3] here we discuss the most relevant features of the results.

$TbMn_2$ crystallizes in the $C15$ cubic Laves phase, the Mn atoms lie at the corner of regular tetrahedra which are packed in the diamond arrangement connected by sharing vertices.[4] The Mn-Mn distance in this compound is very close to that for M-NM instability so that its magnetic properties are extremely sensitive to applied pressure or alloying.

In zero field, below 40 K, antiferromagnetic peaks are detected. This transition is accompanied by a large magnetovolume anomaly ($\Delta V/V=1.5\%$),[5] indicating a large Mn moment of about $2.7\mu_B$. This S_1 structure is generated by the propagation vector $\mathbf{K}_1=(\frac{2}{3},\frac{2}{3},0)$. NMR measurements show that all the Mn sites are magnetic with equal moments.[6] However, the magnetic-moment arrangement within the unitary magnetic cell has not yet been solved. Work is in progress to check whether an earlier proposed structure[7] is the correct one.

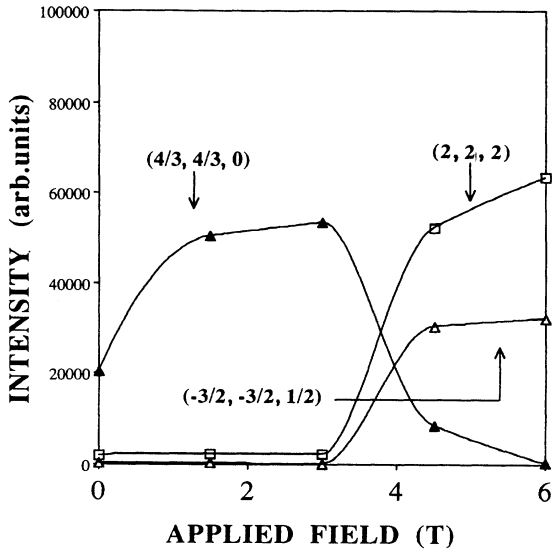

FIG. 2. Field dependence of the reflection peaks at 25 K. $(\frac{4}{3},\frac{4}{3},0)$ and $(-\frac{3}{2},-\frac{3}{2},\frac{1}{2})$ peaks are characteristic of S_1 and S_2 structure, respectively, whereas the (2,2,2) reflection is solely characteristic of the ferromagnetic component of the Mn sublattice.

In the finite field other antiferromagnetic peaks corresponding to a S_2 structure are detected. They can be indexed with a propagation vector $\mathbf{K}_2=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$.

Figure 2 shows the intensity variation of the two antiferromagnetic peaks characteristic of $\mathbf{K}_1$ and $\mathbf{K}_2$ propagation vectors with increasing field, at 25 K. The intensity of the ferromagnetic component of the Mn lattice is also shown. This gives a direct measurement of the relative stability of the two magnetic structures S_1 and S_2. At a critical field $h_c=4.5$ T a transition from S_1 to S_2 is observed, which is also reflected in the ferromagnetic component of the Mn lattice. At lower temperature this criti-

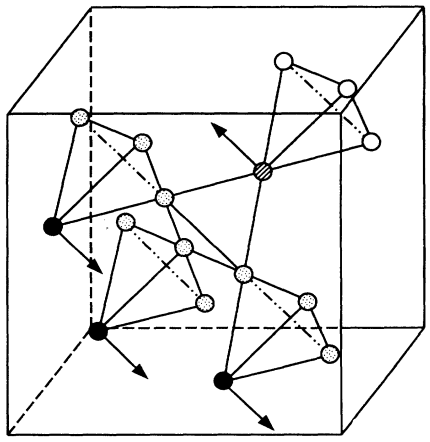

FIG. 3. Antiferromagnetic component of the S_2 structure (for clarity only the Mn sites are shown): ○ and ○ are nonmagnetic sites belonging to different (111) planes, ● and ○ represent magnetic Mn sites. We do not show the ferromagnetic component of the magnetic sites which is along the [111] axis.

cal field increases.

In the S_2 structure, analysis of the magnetic structure factors of the antiferromagnetic peaks and especially the fact that $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ is relatively stronger than $(\frac{3}{2},\frac{3}{2},\frac{3}{2})$ and $(\frac{5}{2},\frac{1}{2},\frac{1}{2})$ requires that the Tb atoms at $\frac{1}{8}\frac{1}{8}\frac{1}{8}$ and $-\frac{1}{8}-\frac{1}{8}-\frac{1}{8}$ are coupled antiferromagnetically, parallel, and antiparallel, respectively, to the Mn moment at $\frac{1}{2}\frac{1}{2}\frac{1}{2}$. This implies that the center of symmetry at the origin is combined with time inversion. This time inversion symmetry together with the propagation vector $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ is not consistent with the existence of moment on the other three Mn atoms of the Mn tetrahedra, which are at the centers of symmetry which like that at the origin are combined with time reversal. However, it is not possible to distinguish between paramagnetic or nonmagnetic sites. The same S_2 structure has been observed by powder neutron diffraction in $Tb_{1-x}Sc_xMn_2$ ($x=0.03$).[8] For this compound NMR measurements[9] have indeed proved the coexistence of magnetic and nonmagnetic sites.

Figure 3 shows the magnetic structure of the Mn sublattice of $TbMn_2$, corresponding to the S_2 state. In this mixed phase only 25% of the Mn sites are magnetic. The same S_2 structure is obtained for a Mn-Mn distance just below a critical value $d_c \approx 2.7$ A,[5] as has been shown by neutron experiments on $Tb(Mn_{0.96}Fe_{0.04})_2$ (Ref. 3) and $TbMn_2$ under pressure.[10] The S_2 structure is also observed in $DyMn_2$ where the Mn-Mn distance is very close to the critical value, and both neutron[11] and NMR (Ref. 6) experiments indicate this same mixed structure.

To summarize, this neutron experiment on $TbMn_2$ confirms that the mixed ordered phase can be stabilized by the application of a magnetic field, showing the soundness of the proposed model in which frustration and instability of the magnetic moment control the behavior of the RMn_2 systems.

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[1]For a recent review on frustrated systems, see P. Chandra and P. Coleman, in *Proceedings of Ecole d'été de physique théorique: Strongly interacting fermions and high-T_c superconductivity,* Les Houches Summer School Proceedings Session LVI, edited by J. Zinn-Justin and B. Douçot (North-Holland, Amsterdam, in press).

[2]R. Ballou, C. Lacroix, and M. D. Nunez Regueiro, Phys. Rev. Lett. **66**, 1910 (1991).

[3]P. J. Brown, B. Ouladdiaf, R. Ballou, J. Deportes, and A. S. Markosyan (unpublished).

[4]K. Nassu, L. V. Cherry, and W. E. Wallace, J. Phys. Chem. Solids **16**, 123 (1960).

[5]H. Wada, H. Nakamura, Y. Yoshimura, M. Shiga, and Y. Nakamura, J. Magn. Magn. Mater. **70**, 134 (1987).

[6]K. Yoshimura, M. Shiga, and Y. Nakamura, J. Phys. Soc. Jpn. **55**, 3585 (1986).

[7]M. L. Corliss and M. J. Hastings, J. Appl. Phys. **35**, 1501 (1964).

[8]R. Ballou, R. M. Galera, E. Lelievre, and B. Ouladdiaf (unpublished).

[9]M. Shiga, J. Hirokawa, H. Wada, and Y. Nakamura, J. Phys. Soc. Jpn **59**, 1410 (1990).

[10]S. Mondal, R. Cywinsky, S. H. Kilcoyne, B. D. Rainford, and C. Ritter (unpublished).

[11]C. Ritter, S. H. Kilcoyne, and R. Cywinski, J. Phys. Condens. Matter **3**, 727 (1991).