First-order antiferromagnetic phase transition in MnS_2†

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(Received 15 April 1976)

The antiferromagnetic phase transition in MnS_2 has been found to be first order in character. This is discussed in relation to recent theoretical analyses of Mukamel, Krinsky, and Bak.

It was recently pointed out by Mukamel[1] that certain phase transitions that involve a doubling of the unit cell, are described by n-component vector models with $n \geq 4$. The critical properties of such a structure transition in NbO_2, for which the order parameter is four dimensional, were studied by the the renormalization-group method. In a second paper,[2] Bak, Krinsky, and Mukamel extended this analysis to first-order transitions. They remarked that within the framework of the Landau-Lifshitz theory, transitions are expected to be first order on general grounds if the symmetry properties of the order parameter have certain known characteristics, or on particular grounds if coefficients in the free-energy expansion have special values. They then proposed that, in addition to these criteria, transitions may be expected to be first order in nature if the Landau-Ginsburg-Wilson Hamiltonian does not possess a stable fixed point in $4-\epsilon$ dimensions.

In a later series of papers,[3-5] Mukamel, Krinsky, and Bak examined a number of antiferromagnetic transitions and showed, in particular, that MnS_2 belongs to a class of $n=6$ vector models having a unique stable fixed point. They computed the critical exponents corresponding to this fixed point to $O(\epsilon^2)$.

We undertook a determination of the critical index β, characterizing the staggered magnetization, in MnS_2, in order to check the specific prediction of the theory, but found, in fact, that the transition is first order. The observed first-order behavior cannot be explained by purely symmetry considerations, but must be dependent upon the particular values of the coefficients in the Hamiltonian describing MnS_2. Although symmetry would allow a second-order transition, the particular Hamiltonian corresponding to MnS_2 must lie outside the region of parameter space from which it can, by the renormalization-group transformation, approach the stable fixed point. This is in contrast with the situation in MnO, UO_2, Cr, and Eu for which the effective Hamiltonians have no stable fixed points. For these systems, the renormalization-group calculations allow a prediction[2] of the observed first-order behavior from symmetry considerations alone.

MnS_2 is cubic above the Néel point, with space group $Pa3$, and consists of an NaCl-like arrangement of Mn^{+2} and $(S_2)^{2-}$ ions, with the axes of the $(S_2)^{2-}$ groups directed along the various body dia-

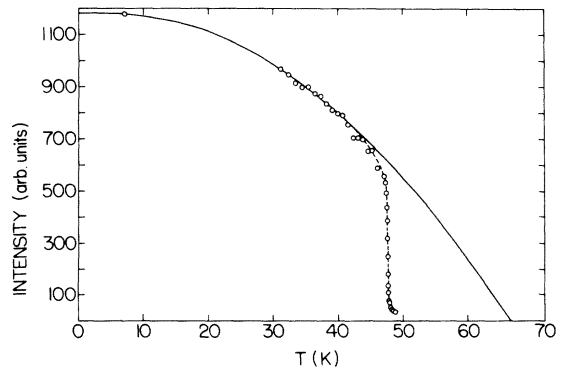

FIG. 1. Temperature dependence of the intensity of the $(\frac{1}{2}01)$ antiferromagnetic reflection. Solid curve is the Brillouin function for $S=\frac{5}{2}$.

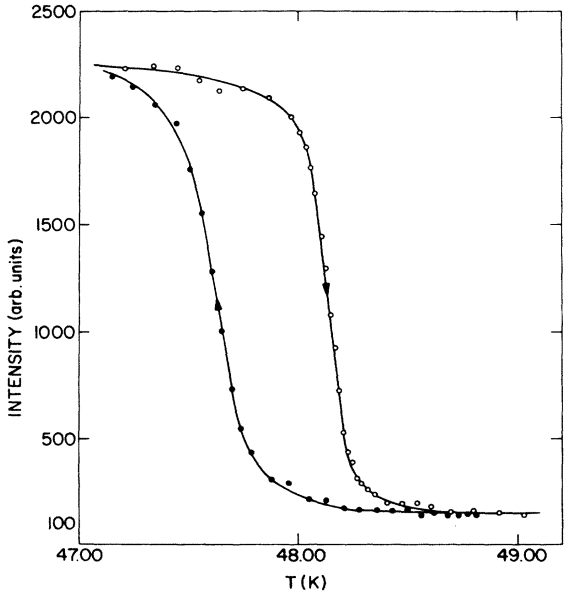

FIG. 2. Hysteresis of the antiferromagnetic transition. Arrows indicate direction of temperature variation.

gonals of the cell. The magnetic structure[6] is ordering of the "third" kind and is described by a propagation vector $\vec{k} = (\frac{1}{2}, 0, 1)2\pi/a_0$, where a_0 is the chemical cell constant. The magnetic unit cell is thus doubled along the x axis. Planes perpendicular to this axis are individually antiferromagnetic and alternate with a period equal to the chemical cell length. The spin orientation is parallel to the doubled direction (x axis).

The material used in this investigation was very pure, polycrystalline, natural hauerite from Raddusa, Sicily. It is the same specimen previously used in the determination of the magnetic structure. The intensity of the $(\frac{1}{2}0\,1)$ antiferromagnetic reflection was monitored as a function of the temperature, which was allowed to drift at approximately 0.1 °K/min, a rate considerably slower than that required for thermal equilibration. A typical run is shown in Fig. 1, in which the Brillouin function for spin $\frac{5}{2}$ is included as an indication of the temperature dependence expected for a second-order transition. Figure 2 shows the hysteresis observed on cycling through the transition. This behavior is reproducible on repeated cycling.

ACKNOWLEDGMENTS

We wish to thank D. Mukamel, P. Bak, and S. Krinsky for very useful discussions.

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† Research performed under the auspices of the U. S. Energy Research and Development Administration.

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