NEUTRON-SCATTERING EXPERIMENTS ON THE TWO-DIMENSIONAL EASY-PLANE ANTIFERROMAGNET K_2FeF_4

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Spin-wave dispersion and sublattice magnetization of K_2FeF_4 have been examined by neutron scattering and analyzed in terms of renormalized spin-wave theory appropriate for the easy-plane square lattice. Numerical results are $J = (-11.5 \pm 0.6)\ \mathrm{cm}^{-1}$, $D = (3.6 \pm 0.5)\ \mathrm{cm}^{-1}$, and $E(T = 0) = (-0.37 \pm 0.10)\ \mathrm{cm}^{-1}$. Near T_N the sublattice magnetization follows a power law, with $T_N = (62.7 \pm 0.6)$ K and $\beta = 0.13 \pm 0.01$.

The main objective of the present investigation is to determine the spin-wave dispersion and sublattice magnetization in the two-dimensional (2D) easy-plane antiferromagnet K_2FeF_4. Accordingly, we performed both elastic and inelastic neutron scattering in the temperature range 4.2–72 K with a triple-axis spectrometer at the HFR reactor in Petten. In fig. 1 the results are collected of a number of constant-$\boldsymbol{Q}$ scans along the [100] direction in the magnetic Brillouin zone. Two distinct magnon branches could be tracked up to $\zeta_x = ak_x/2\pi \approx 0.2$. Further experiments showed the energy of both branches to be independent of k_z, i.e. the spin waves are truly 2D.

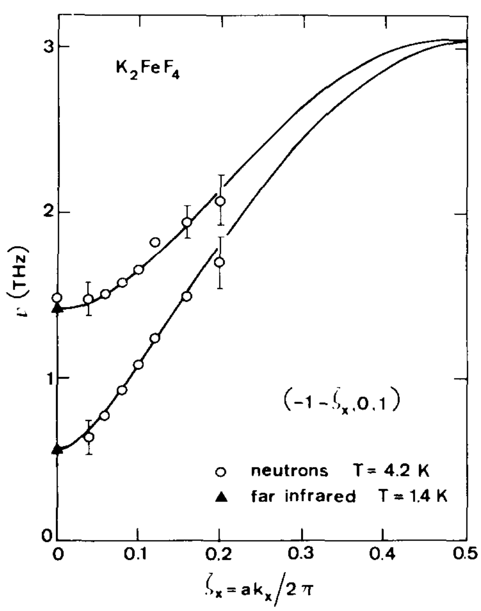

Fig. 1. Spin-wave dispersion at 4.2 K versus k_x, with k_y=0. The solid curve represents spin-wave theory. The black triangles indicate supplementary results of far-infrared spectroscopy.

To calculate the spin-wave energies we assume the Hamiltonian

$$\mathcal{H} = |J| \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \sum_i \left[D S_i^{z2} + E\left(S_i^{x2} - S_i^{y2}\right)\right], \tag{1}$$

in the usual notation; $D(>0)$ represents the out-of-plane anisotropy and E the smaller in-plane anisotropy. Holstein–Primakoff transformation of eq. (1), followed by the introduction of spin waves and diagonalization, leads to two spin-wave branches, whose energies are to lowest order in $1/2S$ given by

$$h\nu_{\pm}(\boldsymbol{k}) = 4|J|S\left\{\left[1 + (D - 3E)/4|J|\right]^2 - \left[\gamma_{\boldsymbol{k}} \pm (D + E)/4|J|\right]^2\right\}^{1/2}. \tag{2}$$

Here, $\gamma_{\boldsymbol{k}} = \cos\frac{1}{2}k_x a \cos\frac{1}{2}k_y a$. The ν_+ modes, having the lowest energy, correspond to mainly in-plane spin deviations; the ν_- modes to mainly out-of-plane deviations. A best fit is made of eq. (2) to the data, with the results $J = (-11.7 \pm 0.6)\ \mathrm{cm}^{-1}$, $D = (3.0 \pm 0.5)\ \mathrm{cm}^{-1}$, and $E = (-0.20 \pm 0.05)\ \mathrm{cm}^{-1}$. It is represented by the solid curves in fig.1, which are in excellent agreement with experiments. In a more precise calculation, eq. (2) is extended to include corrections up to first order in $1/2S$. The first-order renormalized eq. (2) yields a dispersion which only marginally deviates from the solid curves, but the spin-wave parameters are modified to $J = (-11.5 \pm 0.10)\ \mathrm{cm}^{-1}$, $D = (3.6 \pm 0.5)\ \mathrm{cm}^{-1}$, and $E = (-0.37 \pm 0.10)\ \mathrm{cm}^{-1}$.

To determine the variation with temperature of the sublattice magnetization M the intensity of the magnetic $(\bar{1}00)$ reflection was recorded (fig. 2). In the low-temperature regime, $M(T)$ has also been calculated from the above spin-wave formalism including temperature-dependent renormalization (solid curve). Good agreement is obtained up to ~50 K. Here, the result of J, D, and E just obtained have been inserted. It has been assumed that J and D are constant, and E scales with the asymmetry parameter of the field gradient derived from Mössbauer experiments [1]. These assumptions also lead to a correct description of the observed temperature variation of the spin-wave energies. For comparison, in fig. 2 also the result is given, as the dashed line, of the same calculation but with both D and E taken independent of temperature. To demonstrate the improvement obtained by renormalization, the result of an unrenormalized calculation is finally given as the dashed-dotted line, which departs from experiment beyond ~30 K.

In the critical regime the sublattice magnetization displays a well-defined transition, going to zero at $T_N = (62.7 \pm 0.6)$ K according to a single power law with exponent $\beta = 0.13 \pm 0.01$. The value for β is to be compared with Onsager's result $\beta = \frac{1}{8}$ for the 2D Ising system. It should be noted that T_N is confirmed by critical-scattering experiments not discussed here. The results in the critical regime, however, deviate from those of the Mössbauer work [2], where a large transition region from 63 to 70 K was found. In Rb_2FeF_4, Wertheim et al. [3] have reported similar effects. Possibly, residual short-range order within the a–b planes still shows up in the Mössbauer spectra somewhat above T_N. As a matter of fact, 2D critical scattering, which essentially reflects short-range order, is observed to persist in the same temperature regime.

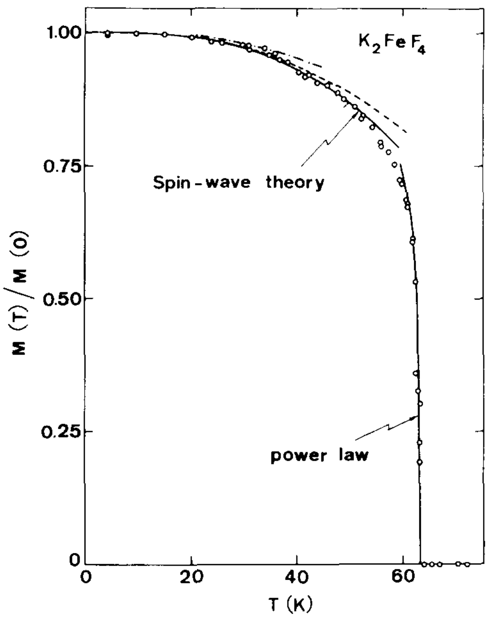

Fig. 2. Normalized sublattice magnetization versus temperature.

References

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