# Direct observation of low-frequency magnetic fluctuations of 2*D* magnetic materials near the ordering temperature

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A magnetic noise in the frequency band 0.01–10 Hz has been observed with the help of a SQUID magnetometer in a zero field in the 2*D* magnetic materials $(CH_3NH_3)_2 \cdot CuCl_4$ and $Co(HCOO)_2 \cdot 2H_2O$ near the transition temperature. In the case of the Ising antiferromagnet $Co(HCOO)_2 \cdot 2H_2O$ the fluctuations reach a maximum at $T_N$, while in the Heisenberg ferromagnet $(CH_3NH_3)_2 \cdot CuCl_4$ they reach a maximum 0.4 K higher, apparently because a Kosterlitz-Thouless topological transition occurs in such systems. In both magnetic materials the noise has a $1/f$ spectrum.

Critical fluctuations in order parameters are an important characteristic of second-order transitions. In magnetic systems they are usually detected from the frequency dependence of the susceptibility or from neutron experiments. The critical dynamics of magnetic materials depends strongly on the structure of the material and reflects the nonlinear properties of magnetic materials. Only recently has active research on this dynamics begun.

To the best of our knowledge, direct measurements of magnetic fluctuations have previous been carried out for only two cases: 1) They have been carried out in several spin glasses,[1] in which a $1/f$ singularity appears in the spin correlation function by virtue of the logarithmic frequency dependence of the susceptibility, and a $1/f$ noise correspondingly appears in the spectral density of fluctuations. 2) In 1985, Hahn *et al.*[2] detected temperature-independent fluctuations of the spins of $^{35}Cl$ nuclei in $NaClO_3$ at the frequency of the nuclear quadrupole resonance.

In the present letter we are reporting measurements of the spectrum of magnetic fluctuations in a zero field by means of a SQUID magnetometer over the frequency band 0.01–10 Hz. The receiving coil of the superconducting magnetic-flux transformer had a coaxial design. The magnetometer feedback signal $U(t)$, which was proportional to the induced magnetic flux, was measured with a Shch300 digital voltmeter, and its readings were stored in a DVK-3 computer at a frequency of 20 Hz. A Fourier analysis of the function $U(t)$ was then carried out, and the measurements were repeated 10 times at each temperature point. The resulting values of the squares of the Fourier coefficients were averaged. The noise of the empty magnetometer at the frequency 1 Hz corresponded to a flux of $1.5 \times 10^{-4}$ $\Phi_0/Hz^{1/2}$. The sample was inside a copper cup with a wall temperature of 0.1 mm. A heater with a bifilar winding was in the lower part of this cup. The cup temperature was measured by an Au:Fe–Cu thermocouple with respect to the temperature of liquid helium and was held constant within 0.05 K.

The $2D$ antiferromagnet $\mathrm{Co(HCOO)_2 \cdot 2H_2O}$. This sample was a polycrystalline powder weighing 160 mg, which filled the copper cup. The magnetic properties of this material correspond to the Ising model with $d = 2$. The crystal structure is monoclinic. The magnetic cobalt ion is parallel to the (100) plane. The Néel temperature is 5.1 K. The anisotropy of the $g$-factor is $g_{\parallel}/g_{\perp} \approx 3$ (Refs. 3 and 4). Figure 1a shows values of the noise spectral density in a unit interval (1 Hz), averaged over the three frequency intervals 0.01–0.1, 0.1–1, and 1–10 Hz. We see that in a narrow region on the order of 0.1 K wide, near the phase transition temperature the fluctuations in the frequency interval 0.01–1 Hz increase. It is natural to link the observed fluctuations with critical fluctuations, although the sharp increase in these fluctuations in the direction of ultralow frequencies is noteworthy (Fig. 2b).

The $2D$ ferromagnet $\mathrm{(CH_3NH_3)_2 \cdot CuCl_4}$. This ferromagnetic material is a member of a group of $2D$ magnetic materials which have the $\mathrm{K_2NiF_4}$ structure. It is a Heisenberg ferromagnet with a weak $XY$ anisotropy; the interaction between layers is $10^{-4}$ of the exchange interaction in the plane; and the Curie temperature is 8.9 K (Ref. 5). The samples were thin wafers $\sim 3 \times 4 \times 0.2$ mm in size with a total weight of 8 mg. We measured the fluctuations of the moment in the plane, $M_{\parallel}$ (the axis of the receiving coil ran parallel to the ordering planes), and in the perpendicular direction, $M_{\perp}$. The results are qualitatively different from the results found previously (Fig. 1b). In this case the fluctuations in the moments in the plane ($M_{\parallel}$) reach a maximum about 0.4 K above $T_c$, although the frequency dependence is similar in the two cases (Fig. 2b). The power of the fluctuations in the perpendicular component, $M_{\perp}$, is lower than that in the parallel component by a factor of about four; i.e., the predominant arrangement of the spins in the plane is clearly expressed. Furthermore, there are two maxima: one at $T_c$ and another coinciding with the maximum in the fluctuations in

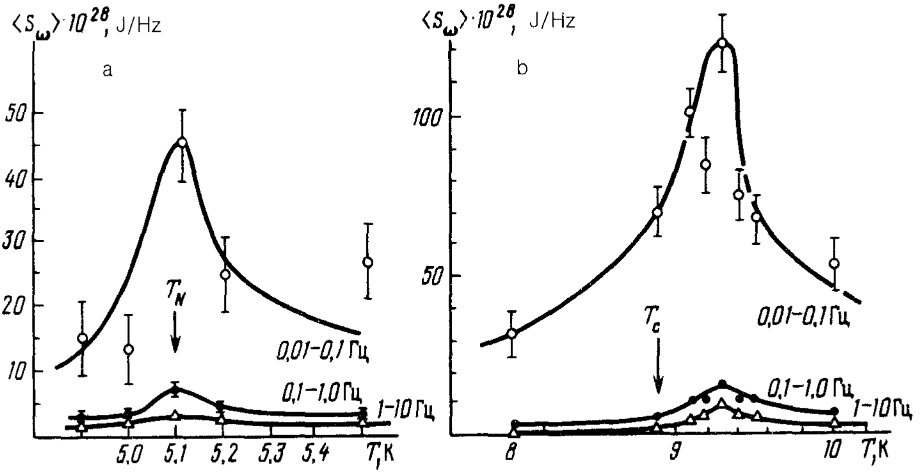

FIG. 1. Temperature dependence of the average noise spectral density in three frequency intervals: 0.01–0.1, 0.1–1, and 1–10 Hz. a—polycrystalline $\mathrm{Co(HCOO)_2 \cdot 2H_2O}$; b—$\mathrm{(CH_3NH_3)_2 \cdot CuCl_4}$. The easy plane of the crystal is perpendicular to the plane of the turns of the receiving coils of the flux transformer.

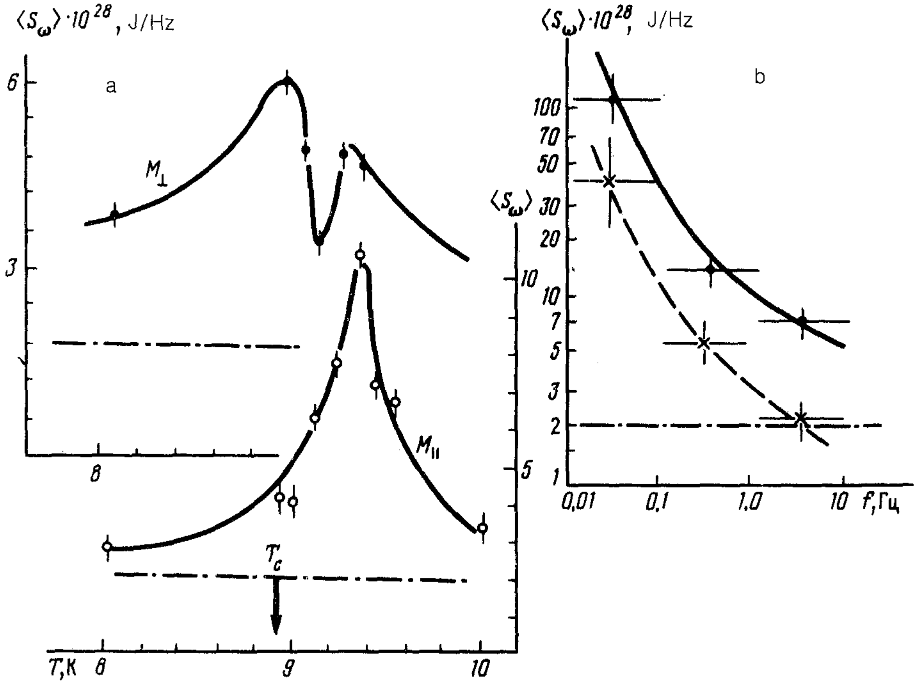

FIG. 2. a—Temperature dependence of the spectral density of the magnetic fluctuations, averaged over the band 0.01–10 Hz, for two orientations of the $(CH_3NH_3)_2\cdot CuCl_4$ crystals: $M_\parallel$ and $M_\perp$; b—spectrum of magnetic fluctuations. ×) $Co(HCOO)_2\cdot 2H_2O$ ($T = 5.1$ K); ●) $(CH_3NH_3)_2\cdot CuCl_4$ ($T = 9.3$ K). The dot-dashed line is the noise level of the empty magnetometer.

$M_\parallel$. This situation is illustrated clearly by Fig. 2a, which shows for comparison the temperature dependence of the power levels of both the $M_\parallel$ and $M_\perp$ fluctuations, averaged over the entire frequency band studied, 0.01–10 Hz.

The low-frequency magnetic noise observed here is undoubtedly related to fluctuation phenomena in the critical region. However, this noise in the case of 2*D* Ising systems is qualitatively different from that in easy-plane Heisenberg systems: In the Ising case the fluctuations are amplified directly in the region of the temperature-induced phase transition, while in the easy-plane Heisenberg ferromagnet they reach a maximum above the Curie temperature. According to the theoretical understanding, some unusual excitations—magnetic vortices—should exist in the 2*D XY* magnetic materials. A dissociation of vortex pairs initiates a phase transition, a so-called (Berezinskiĭ-) Kosterlitz-Thouless transition.[6] This transition should occur earlier along the temperature scale than that caused by spin waves. Nevertheless, the influence of spin-wave excitations on the disruption of the order in the planes should be manifested in some way. It might be suggested that the maximum in the magnetic fluctuations in $(CH_3NH_3)_2\cdot CuCl_4$ is being observed here at the temperature at which the phase transition should have occurred if it had been caused by spin waves and if vortex excitations had not occurred in the system.

The frequency dependence of the fluctuations is similar to the well-known $1/f$ noise in various systems, but the nature of the amplification of these large-scale fluctuations in the critical region is not clear.

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