New intermediate phase in the spiral antiferromagnet $CsCuCl_3$ induced by a strong magnetic field

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A new magnetic phase in the intermediate temperature interval has been discovered in the spiral antiferromagnet $CsCuCl_3$ in a magnetic field $H \gtrsim 50$ kOe. The experimental results are explained on the basis of a weak dipole-dipole interaction.

The antiferromagnetic crystal $CsCuCl_3$ has attracted interest because it has been found to exhibit piezoelectric properties, and an optical activity, and its lattice undergoes a helicoidal distortion[1] upon a structural phase transition (at $T = 423$ K) caused

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a cooperative Jahn-Teller effect.[1)] In the magnetically ordered state, which is reached at $T_N = 10.7$ K, the antiferromagnetic interaction between the chains in $CsCuCl_3$ forms a triangular magnetic structure in the basis plane. The strong exchange interaction within chains, along with the slight anisotropic interaction (described by a Lifshitz invariant), gives rise to a modulated spiral structure along the c axis.[2]

In this letter we report a study of the phase transition to the magnetically ordered state in $CsCuCl_3$ in a strong magnetic field. For the measurements we use a null-balance magnetometer with a superconducting solenoid in fields up to 80 kOe over the temperature interval 2–24 K.

The temperature dependence of the magnetic susceptibility in weak fields corresponds to a spiral ordering of the spins in $CsCuCl_3$, and the values of the constants for intrachain and interchain exchange are $J_1/k = 25$ K and $J_2/k = -3.9$ K, in agreement with data reported by Tazuke *et al.*[3]

In a field of 3.3 kOe, applied along the c axis, we observe a sharp change in slope on the $M(T)$ curve (curve 1 in Fig. 1). After this change in slope, the magnetic moment decreases with decreasing temperature. An increase in the field changes the shape of the $M(T)$ curve: Specifically, at $H \gtrsim 50$ kOe a sharp peak appears in the vicinity of the phase transition, and it increases in height as the magnetic field is raised (curve 3 and 4 in Fig. 1). At the point $T = T_1$, there is another change in slope on the $M(T)$ curve, but

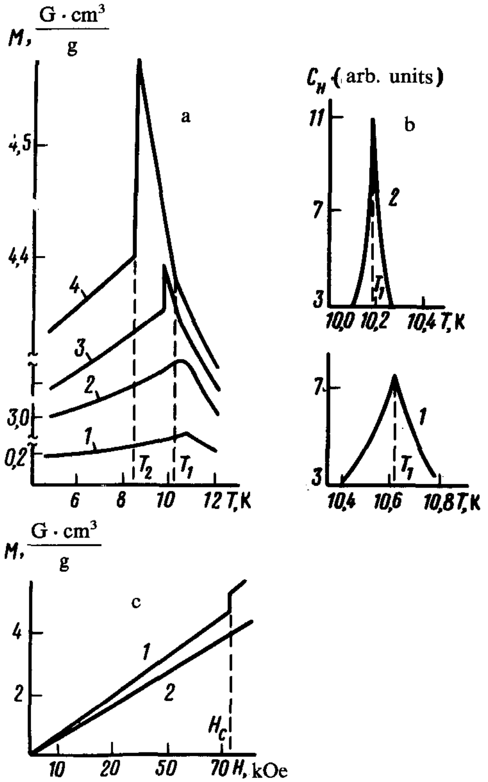

FIG. 1. The magnetic behavior of $CsCuCl_3$ in a field directed along the helicoidal axis ($H \parallel c$). a: Temperature dependence of the magnetic moment of $CsCuCl_3$ for various fields. 1—3.3 kOe; 2—50 kOe; 3—60 kOe; 4—71.6 kOe. b: Magnetic part of the heat capacity, C_H, as a function of the temperature. 1—At $H = 60$ kOe; 2—$H = 71.6$ kOe. c: Dependence of the magnetic moment of $CsCuCl_3$ on the external field. 1—At $T = 8.7$ K; 2—$T = 4.2$ K.

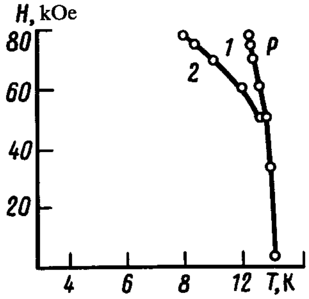

FIG. 2. The H, T phase diagram of $CsCuCl_3$. 1—Region of an incommensurate phase with a modulation period in the basis plane; 2—spiral structure modulated along the c axis; P—paramagnetic phase.

at $T < T_1$ the magnetic moment of the sample initially rises rapidly with decreasing temperature, reaches a value $T = T_2$, and then decreases abruptly. At $T > T_1$, the $M(H)$ curve corresponds to a paramagnetic behavior; at $T > T_1$, a jump in the magnetization appears on the magnetization curve at the field $H = H_c$ (Fig. 1c), implying the formation of a magnetically ordered state. Figure 1b shows curves of d^2M/dT^2, which are proportional to the magnetic component of the heat capacity, C_H, near the points T_1 for two values of H. The point at which the structural feature appears on the C_H curve coincides with the point (T_1) of the change in slope on the $M(T)$ curve. We show below that this change in slope corresponds to the formation of long-period modulations in the basis plane.

We see from the $CsCuCl_3$ phase diagram in Fig. 2 that the temperatures T_1 and T_2 decrease with increasing field; the decrease is more pronounced in the case of T_2.

These results can be explained in a noncontradictory way by taking into account the effect of the dipole-dipole interaction between spins. This interaction is important in a study of phase transitions in triangular antiferromagnets,[4] since, under certain conditions, it may lead to an incommensurate state through a conical-point instability.[5]

To see how a field affects the phase diagram of the compound $CsCuCl_3$ with $S = 1/2$, we use a Landau expansion for the free energy in powers of the expectation values of the spin, $\langle S_i^\alpha \rangle$ (we are retaining terms of up to fourth order):

$$F = \frac{1}{2} \sum_{ij} a_{ij}^{\alpha\beta} \langle S_i^\alpha \rangle \langle S_j^\beta \rangle + \sum_i b \langle S_i^\beta \rangle^4 - \sum_i g\mu_B H^\alpha \langle S_i^\alpha \rangle, \quad (\alpha, \beta = x, y, z). \quad (1)$$

Here the Fourier components $a^{\alpha\beta}(\mathbf{q}) = A^{\alpha\beta}(\mathbf{q}) + T\delta_{\alpha\beta}$ are components of the inverse susceptibility tensor, for which the nonlocal term $A^{\alpha\beta}(\mathbf{q})$ represents the Fourier components of a Hamiltonian consisting of the exchange interactions along a chain and between chains, the Dzyaloshinkiĭ interaction (the interaction constant is $D \approx 5$ K), and the dipole-dipole interaction.

In a zero external field, the wave vector of the modes condensed from the paramagnetic phase is determined from the conditions which minimize the smallest eigenvalue $\lambda_-(\mathbf{q})$ of the Hermitian matrix $A^{\alpha\beta}(\mathbf{q})$. The vector $\mathbf{q} = \mathbf{q}_0 = (4\pi/3a, 0, D/J_1c)$, which achieves this minimum for the function $\lambda_-(\mathbf{q})$, also determines the magnetic structure of $CsCuCl_3$, whose period is incommensurate with the lattice period of only

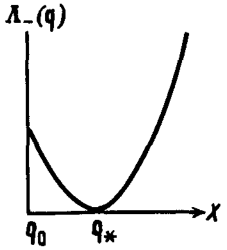

FIG. 3. The smallest eigenvalue, $\Lambda_-(\mathbf{q})$, versus the wave vector $\mathbf{q}$.

the c axis. To take the effect of the field into account, we need to transform from the variables $\langle S_i^\alpha \rangle$ to the slowly varying variables ψ_i^α:

$$\langle S_i^\alpha \rangle = (\psi_i^\alpha \exp(i\mathbf{q}_k \mathbf{r}_i) + \text{ c.c.}) + m^\alpha, \qquad \mathbf{q}_k = \left(\frac{4\pi}{3a}, 0, 0\right) \tag{2}$$

(m^α is the constant component along the field). Substituting (2) into (1), we find that the paramagnetic state becomes unstable with respect to ψ^α as the temperature is lowered. This instability is determined by calculating the eigenvalues of the matrix of coefficients $\tilde{a}^{\alpha\beta}(\mathbf{q})$ of the terms that are quadratic in ψ^α in F. The matrix $\tilde{a}^{\alpha\beta}(\mathbf{q})$ differs from $a^{\alpha\beta}(\mathbf{q})$ in that its elements are renormalized (because of the term of fourth order in ψ^α in the expression for F) by the terms with the factor $m^\alpha m^\beta b$.

If the uniform field H is directed along the axis of the spiral structure, then, beginning at a critical value $H_c \sim [(\gamma_d/J_2)(D/J_1)]^{1/2}$ where $\gamma_d = (g\mu_B)^2/a^3$, the eigenvalues $\Lambda(\mathbf{q})$ of the renormalized matrix $\tilde{a}^{\alpha\beta}(\mathbf{q})$ do not have a minimum at the point $\mathbf{q} = \mathbf{q}_0$ but near it. Figure 3 shows the lowest branch of the function $\Lambda(\mathbf{q}) = \Lambda_-(\mathbf{q})$; the wave vector $\mathbf{q}_*$ corresponding to the minimum of this function is displaced from the symmetric point of the reciprocal lattice, $\mathbf{q}_k$, by an amount proportional to the dipole-dipole interaction. The periods of the modes $\psi_i^\alpha = \varphi_\alpha \exp i\mathbf{q}_1\mathbf{r}_i (\mathbf{q}_1 = \mathbf{q}_* - \mathbf{q}_k)$ that arise now become incommensurate with the lattice constant in the basis plane. The constant component of the magnetization $m(T)$ along the field H is given in this case by

$$m = \begin{cases} \dfrac{H}{\Lambda_-(0) - \Lambda_-(\mathbf{q}_*) - 24b\,|\varphi_x|^2}, & T \leqslant T_1 \quad (T \geqslant T_2); \\[2ex] \dfrac{H}{\Lambda_-(0) + T}, & T \geqslant T_1 \end{cases} \tag{3}$$

where $|\varphi_x|^2 = (T_1 - T)/12b$, and $T_1 = -\Lambda_-(\mathbf{q}_*)$; at $T > T_1$, we have $\varphi_x = 0$ in (3). At the mode condensation point ($T = T_1$), there is accordingly a slope on the curve of the magnetization m. At $T < T_1$, the magnetic moment increases more rapidly than in the paramagnetic phase, $T > T_1$, in agreement with experiment (curves 3 and 4 in Fig. 1a).

Calculating the eigenvectors of the matrix $\tilde{a}^{\alpha\beta}$, and comparing the free energies of the spiral state and those of the state with the incommensurate period in the basis plane, we numerically constructed the phase diagram for $CsCuCl_3$. The results of these calculations for the sample studied agree well with the experimental data (Fig. 2). As is found experimentally, the transition between different states with an incommensurate phase is a first-order transition. The temperature interval in which the new intermediate phase exists is $\Delta T \sim (H - H_c)^2$.

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[1)]The lattice symmetry belongs to space group $P6_122$.

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