Effect of high hydrostatic pressure on the flipping of the magnetic moments of sublattices and on the Néel temperature in the antiferromagnetic single crystal $CuCl_2 \cdot 2H_2O$

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The effect of pressure on the phase diagram of copper chloride dihydrate was investigated. It is shown that the law of corresponding states holds for this crystal. The effect of high hydrostatic pressure on the Néel temperature and on the flipping of the sublattice magnetic moments is studied.

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Flipping of the magnetic moments (FMM) of the sublattices can take place in antiferromagnets places in an external magnetic field. FMM was first observed in copper chloride dihydrate.[1] A study of the resonant properties of the antiferromagnet (AFM) $CuCl_2 \cdot 2H_2O$ at high hydrostatic pressures has revealed a strong dependence of the flipping fields H_n on the hydrostatic compression of the sample.[2]

It is quite urgent to study the FMM under the influence of high hydrostatic pressure and to trace the variation of the Néel temperature and of the field H_n by plotting the phase diagram for the given single crystal. We chose the NMR method,[1,3] and used the spectrometer employed in similar researches.[4] A high hydrostatic pressure, on the order of 10 kbar, was produced in an autonomous chamber in the temperature interval 1.6–5.4 °K. The pressure was measured by determining the temperature of the superconducting transition on the pressure with accuracy ±100 bar. The behavior of the proton-resonance line near the Néel temperature[3] has made it possible to plot $T_N = f(P)$ (Fig. 1). The

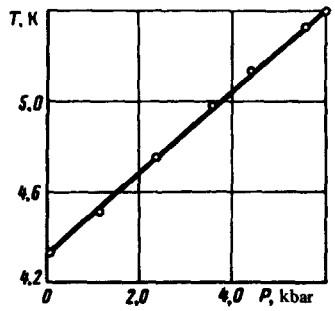

FIG. 1. Pressure dependence of the Néel temperature.

Néel temperature was measured in a 4-kOe external field. The low intensity of the external magnetic field was chosen from considerations of its influence on the transition temperature. This value of the field lies within the temperature-measurement error ± 0.005 °K, and the influence of the magnetic field can be neglected. Measurement yielded $dT_N/dP = 1.6\times10^{-4}$ deg/bar.

We investigated in detail the effect of pressure on the field H_n, and also on the triple point of the phase diagram H_3 (Fig. 2). The value of H_n was measured by two methods, by determining the character of the proton resonance near the easy magnetization axis, [1] and by the NMR signal from the domain walls. At a field orientation along the easy axis, in the zone where the intermediate state was produced under the influence of the external magnetic field, one can see the signal from the domain walls. [4] H_n was determined from the maximum of the intensity. The results obtained by the two methods were in good agreement. The measurement accuracy was ± 1 Oe. The triple point H_3 was determined from the vanishing of the signal from the domain walls.

Figure 3 shows a plot of $H_n/H_3 = f(T/T_3)$. The dependence is quadratic, and the plot itself confirms the law of corresponding states for antiferromagnets, i.e., H_n/H_3 is a function of the ratio T/T_3, where T_3 is the temperature of the triple point on the HT phase diagram. The value of the pressure does not affect the shape of the presented curve.

Owing to the magnetostriction, the external hydrostatic pressure leads to a redefinition of the exchange

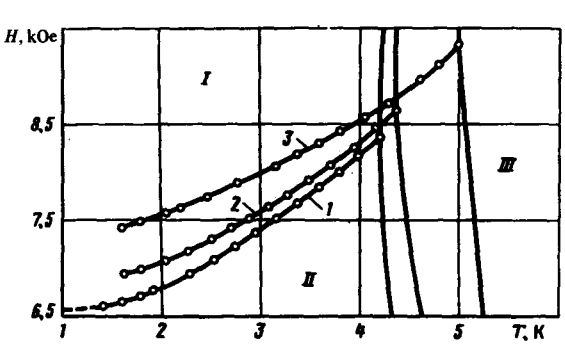

FIG. 2. Phase diagram of $CuCl_2 \cdot H_2O$. Curves 1, 2, and 3 are drawn for $P = 0.2$, 5 kbar, and 3 kbar, respectively. The numbers I, II and III designate the spin-flop, antiferromagnetic, and paramagnetic regions.

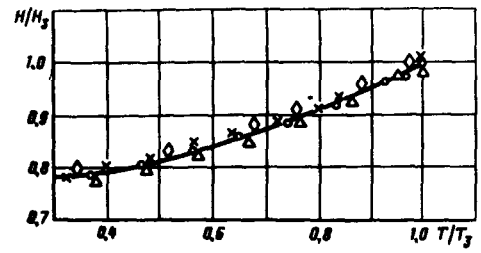

FIG. 3. Diagram of corresponding states: ×—5.3 kbar, ○—2 kbar, Δ—$P = 0$, ◊—10 kbar.

integral and of the anisotropy constants. These renormalizations can be carried out by starting from the following expression for the magnetostriction energy

$$H_m = \lambda^{(1)}_{ik,pq}\, l_i\, l_k u_{pq} + \lambda^{(2)}_{ik,pq}\, m_i\, m_k u_{pq}\ , \tag{1}$$

where $\lambda^{(1)}$ and $\lambda^{(2)}$ are the exchange-magnetostriction constants corresponding to the vectors l and m, while the indices i, p, k, and q serve to denote the projections of these vectors on the axes x, y, and z.

Allowance for the symmetry causes only the components $\lambda_{\alpha\beta}$ ($\alpha, \beta \leq 3$) and λ_{44}, λ_{55}, and λ_{66} (where α and β are the Voigt indices) to be different from zero.

Without dwelling on the calculations, we present an expression for the exchange integral under pressure

$$I(P) = I_0 + [\lambda^{(1)} - \lambda^{(2)}]\frac{\kappa P}{S^2}\ , \tag{2}$$

where κ is the summary compressibility, P is the pressure, S is the spin (equal to 1/2), I is the exchange integral ($I = \sum_i I(R_i)$), and $\lambda^{(1)} \equiv \lambda^{(1)}_{ii,pp}$.

The renormalization of the anisotropy constants K leads to

$$K(P) = K + \kappa P(\lambda_{22} - \lambda^{(1)}). \tag{3}$$

Noting that the transition field H_n and the field H_3 are proportional to $\sqrt{2IK}$, and that $T_3 = bI$ (b is a constant coefficient), we represent the equation for $H_n(P, T)$ in the form

$$H_n(P, T) = H_3(P, 0)\left[1 + \frac{2}{3S^3}\left(\frac{T}{I(P)}\right)^2\right]. \tag{4}$$

The ratio of the transition field to the triple-point field then becomes

$$\frac{H_n(P, T)}{H_3(P)} = \frac{1}{a}\left[1 + b^2\frac{2}{3S^3}\left(\frac{T}{T_3}\right)^2\right]. \tag{5}$$

where a is the constant coefficient in the formula $H_3 = a\sqrt{2IK}$. Formula (5) is a reflection of the law of corresponding states.

The temperatures T_3 and T_N (Fig. 2) satisfy at all pressures the relation $T_3/T_N \approx 0.95$.

From the curve of Fig. 3 and from (5) we obtain

is in this case $I_0 = (T_3/b)_{P=0} = 17.8\,^\circ$K. The value of the exchange integral I_0 can be obtained by using the value of the exchange field $H_e = 1.2\times10^5$ Oe.[5]

Recognizing that $2SI_0 = 2\mu_0 H_e$, where μ_0 is the Bohr magneton, we obtain $I_0 = 17.6\,^\circ$K. These values of I_0 are in good agreement.

From I_0 we can calculate T_N by using different theoretical variants,

	MFT	HTE	CS	TS	EXP
T_N K	8.9	4.45	3.91	3.02	4.33

where MFT, HTE, CS, TS, and EXP stand for the molecular field theory,[6] the high-temperature expansion,[7] the Callen splitting of the Green's function,[8] the Tyablikov splitting of the Green's functions,[7] and the experimental data,[3] respectively.

It is seen from the table that the best agreement with experiment is obtained with the high-temperature expansion.

The results indicate that a high hydrostatic pressure: a) exerts an influence on the field H_n at which the flipping of the magnetic moments of the sublattices takes place in $CuCl_2 \cdot 2H_2O$; b) shift the Néel temperature towards higher values; c) changes linearly the value of the exchange integral.

The law of corresponding states holds in the antiferromagnetic single crystal $CuCl_2 \cdot 2H_2O$.

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