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SPECIFIC HEAT OF ANHYDROUS $CrCl_3$ BETWEEN 4.5 AND 20°K IN A MAGNETIC FIELD

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Anhydrous $CrCl_3$ is a layered antiferromagnet in which the antoferromagnetic ordering sets in at T_c = 16.8°K and two-dimensional ferromagnetism properties are observed [1 - 4].

$CrCl_3$ has a layered crystalline structure of the ($R\bar{3}$) type and, as shown by neutron-diffraction [5], in the antiferromagnetic states the spins are ferromagnetically ordered inside each hexagonal layer of the metallic ions and in neighboring layers separated by Cl ions they are antiparallel and oriented in the basal plane.

The magnetic properties of layered antiferromagnets were first predicted by Landau [6]. Recently a number of authors [7, 8, 2] have considered theoretically the energy spectrum of layered antiferromagnets. They have shown that if the antiferromagnetic interaction between the layers is much weaker than the ferromagnetic interaction in the layer, and if the anisotropy is small, a transition from a three-dimensional antiferromagnet to a two-dimensional ferromagnetic system sets in already at low temperatures.

The changes of the susceptibility of single-crystal $CrCl_3$ [9, 2] have shown that the anisotropy in $CrCl_3$ is weak, the antiferromagnetic coupling between layers is very weak, and the antiferromagnetic order is disturbed in weak fields; the exchange interaction between layers, determined in [2] from the value of $\chi_\perp$, was J_{af}/k = -9.918°K.

Narath and Davis [2] investigated the dependence of the $CrCl_3$ sublattice magnetization on the field (up to 10 kOe) and on the temperature between 0.4 and 8°K, using the method of nuclear magnetic resonance. They estimated the internal field at which the antiferromagnetic ordering is disturbed and above which the spins in the neighboring sublattices become aligned in parallel (ferromagnetic state) at 1.68 kOe, and found $CrCl_3$ to be practically isotropic in the ferromagnetic state. The temperature dependence of the sublattice magnetization in the absence of a field is attributed by Narath and Davis to singularities of the energy spectrum of the two-dimensional ferromagnetic system, with allowance also for the spin-wave interaction. According to their estimates, the ferromagnetic interaction in the layer is J_f/k = 5.25°K and the effective anisotropy field in the antiferromagnetic state is H_A = 650 Oe.

There are known measurements of the specific heat of anhydrous $CrCl_3$ in the absence of a field above 12°K [10, 11]. In these investigations the temperature was of the transition from the paramagnetic to the antiferromagnetic state of $CrCl_3$ was found to be T_c = 16.8°K.

Low-temperature measurements of the specific heat of $CrCl_3$ [3, 4] without a field were made from 2 to 20°K and made it possible to separate the linear term of the magnetic specific heat; this term is typical of a two-dimensional ferromagnetic system. It was found that the magnetic specific heat of $CrCl_3$ between 2 and 8°K is equal to 0.0535T cal/mole-deg. This led to an estimate J_f/k = 5.61°K of the exchange ferromagnetic interaction in the layer, which is close to the result of Narath and Davis.

We have investigated the behavior of the specific heat of $CrCl_3$ in a magnetic field and measured the specific heat in fields up to 8 kOe in the temperature region from 4.5 to 20°K.

We used for the measurements powdered $CrCl_3$, since, as indicated above, the anisotropy in $CrCl_3$ is quite small. A sample of powdered $CrCl_3$ in the form of a cylinder, weighing ∿30 g, was placed in a calorimeter in a transverse magnetic field produced by an electromagnet. The demagnetization field was 15 - 20% of the external field (the sample demagnetization factor was N ∿ 1/2); to estimate the demagnetization field we used the data of Bizette and Terrier [9] on the magnetization of $CrCl_3$. The calorimeter with the sample contained an Allen Bradley carbon thermometer and a heater, and was filled with a small amount of heat-exchange helium. The procedure of the calorimetric measurements was analogous to that described earlier [12, 13].

The results of our measurements of the specific heat of anhydrous $CrCl_3$ in external fields 870, 5800, and 8100 Oe are plotted in Fig. 1 in the coordinates C and T, together with earlier data [4] obtained without a field. We see that the data on the specific heat of $CrCl_3$ without a field and in fields up to ∿8 kOe are close in the temperature range from 4.5 to 12°K, and the specific heat depends on the field above 12°.

Figure 2 shows the same results below 10°K, plotted in the coordinates C/T and T^2, as well as the data without a field at helium temperatures [3]. The points in the presence of a magnetic field fit almost the same line as the points without a field

$$C[\text{cal/mol-deg}] = 0{,}0535\,T + 5{,}11\cdot 10^{-4}\,T^3,$$

where the cubic term describes the specific heat of the lattice, and the linear term the magnetic specific heat. The linear temperature dependence of the magnetic specific heat, which is characteristic of a two-dimensional ferromagnetic system, remains in force also in the presence of a field between 4.5 and 8°K.

It is seen from the results of Fig. 1 that in a field of 870 Oe the specific heat already becomes dependent on the field; the specific heat at the maximum has become smaller in this case. The maximum of the specific heat remained practically unshifted when the temperature was changed in fields 870 and 5800 Oe,

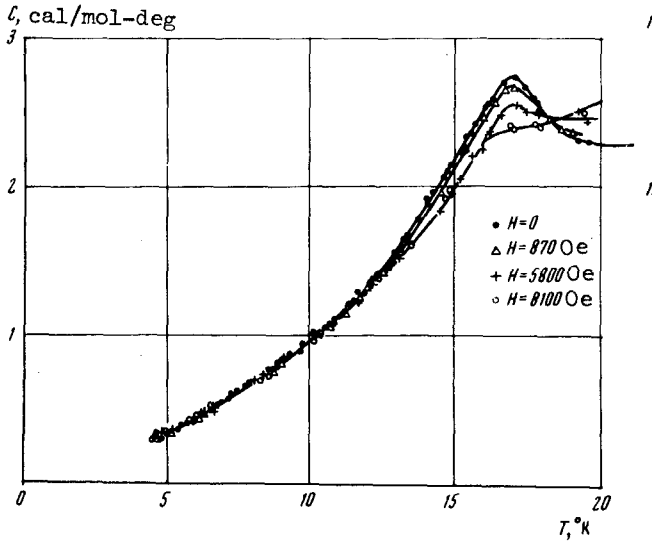

Fig. 1

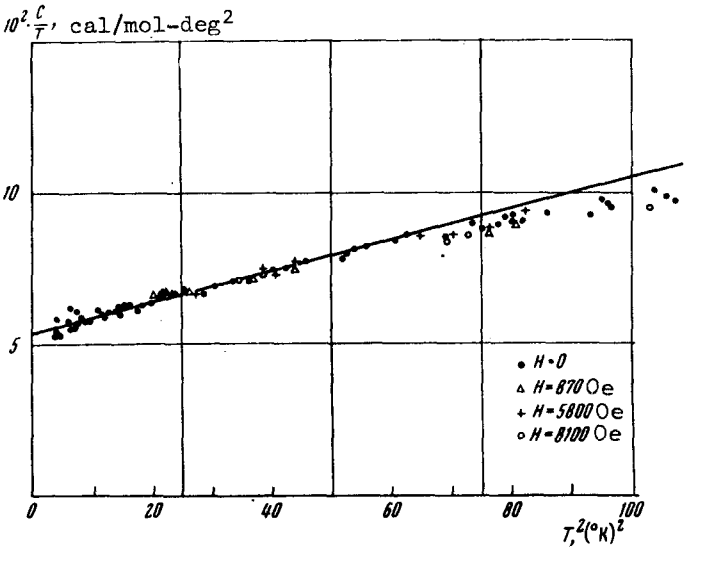

Fig. 2

but the hump on the specific-heat curve became noticeably smaller at 5800 Oe. At 8100 Oe, the specific heat curve becomes even smoother and the maximum disappears. Thus, when a magnetic field is applied the anomaly of the specific heat in $CrCl_3$ becomes gradually smoothed out, starting with weak fields.

The specific-heat peak vanishes in ordinary ferromagnets in a magnetic field [14] because they have no phase transition in a magnetic field. The smoothing of the maximum of the $CrCl_3$ specific heat maximum in a field is apparently also an indication that the phase transition vanishes in the field. This seems to be an inherent property of layered antiferromagnets with weak antiferromagnetic interaction between layers, in which a ferromagnetic ordering sets in already in weak fields.

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METAMAGNETISM IN THE UAs_ZSe_{1-Z} SYSTEM

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The magnetic properties of solid solutions of uranium monocompounds with elements of groups V and VI, of type UX_ZY_{1-Z}, where X = P, As, Sb, Bi, and Y = S, Se, or Te, have been intensively investigated of late. The reason is that the compounds UX have a collinear antiferromagnetic structure, the compounds UY are collinear ferromagnets, and different collinear and noncollinear magnetic structures are observed in pseudobinary solid solutions of these monocompounds in one another [1 - 5]. Thus, for example, in the system UAs_ZSe_{1-Z} in a zero field, at least seven different magnetic structures were observed [3]. Compounds with $0 \leq Z \leq 0.65$ have a collinear ferromagnetic structure; at $0.65 \leq Z \leq 0.7$ noncollinear antiferromagnetic structures that can be described as a sinusoidal modulation of the Z-component of the magnetic moment are observed, and at $0.7 \leq Z \leq 1$ various types of collinear antiferromagnetic structure are