Specific heat of anhydrous $NiCl_2$ between 2 and 30°K

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The specific heat of $NiCl_2$ is investigated at low temperatures between 2 and 30° K. Some features of the temperature dependence of the magnetic specific heat of $NiCl_2$, which are related to the transition to two-dimensional ferromagnetism are discussed, viz., the T^2 dependence at helium temperatures and the region of linear variation with temperature above 14° K. The results are compared with the predictions of the spin-wave theory.

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Investigations of the heat capacity of $NiCl_2$ at helium temperatures[1] indicate that this material has two-dimentional properties. To assess the singularities of the magnetic energy spectrum of $NiCl_2$, we have refined in the present study the measurements of the heat capacity (between 4 and 15°K) and continued them to 30°K. This has made possible a comparison of experiment with theory.

The heat capacity of $NiCl_2$ was measured earlier in the high-temperature region.[2,3] The temperature of the antiferromagnetic transformation was found to be $T_N = 52.3$°K.[3]

The $NiCl_2$ crystal with layered structure of the $CdCl_2$ type has in the antiferromagnetic state an anisotropy of the easy plane type, with a ferromagnetic interaction A between the metallic ions inside each layer, and an antiferromagnetic interaction B between the layers.

The magnetic properties of layered antiferromagnets were first considered by Landau.[4] Recently, a theoretical study was made of the energy spectrum of layered antiferromagnets with easy plane anisotropy, and Yoshimori's results[5] on the temperature dependence of the magnetic heat capacity were used by us in the discussion of the experimental data. It follows from the calculations that if A is significantly larger than B and the anisotropy is low, as is the case with $NiCl_2$, then even at low temperatures a transition takes place to a two-dimensional ferromagnetic system; the T^3 for the magnetic heat capacity then turns into a linear relation.

The table lists the equalized results of our measurements of the heat capacity of $NiCl_2$ between 1.8 and 30°K. In Fig. 1, the results are plotted in the coordinates C/T and T.

At helium temperatures, the dependence of the heat capacity of $NiCl_2$ on the temperature is close to quadratic (dashed line in Fig. 1), and the magnetic contribution to the heat capacity is 3–5 times larger than that of the lattice which will be determined below. It was assumed in this connection[1] that the T^2 dependence for the heat capacity seems to reflect a singularity in the law of spin-wave dispersion in the region of the transition from a three-dimensional antiferromagnet to a two-dimensional ferromagnetic system.

T, °K	C*	T, °K	C	T, °K	C
2.0	0.0110	6.5	0.114	16	0.549
2.25	0.0143	7.0	0.129	17	0.613
2.5	0.0181	7.5	0.145	18	0.679
2.75	0.0223	8.0	0.163	19	0.745
3.0	0.0269	8.5	0.181	20	0.813
3.25	0.0316	9.0	0.200	21	0.884
3.5	0.0365	9.5	0.220	22	0.959
3.75	0.0416	10.0	0.241	23	1.04
4.0	0.0470	11	0.283	24	1.12
4.5	0.0586	12	0.329	25	1.21
5.0	0.0713	13	0.378	26	1.30
5.5	0.0845	14	0.431	27	1.399
6.0	0.0985	15	0.488	28	1.505
				30	1.72

*The heat capacity is given in units of cal/mole-deg.

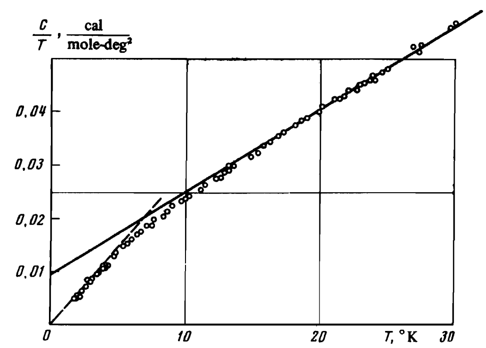

FIG. 1

Above 14°K, the points on Fig. 1 fit a straight line that describes the heat capacity of $NiCl_2$ by the relation

$$C[\text{cal/mole-deg}] = 1.55\cdot 10^{-3}\,T^2 + 0.0098\,T.$$

It can be assumed that the linear term in this relation represents the magnetic contribution characteristic of a two-dimensional ferromagnet, while the quadratic term represents the heat capacity of the lattice.

A quadratic temperature dependence of the heat capacity of the lattice was observed in several nonmagnetic layered halides near hydrogen temperatures, for example in $CdCl_2$.[6] The transition from the T^2 law to the linear law for such layered lattices was discussed theoretically by I. M. Lifshitz;[7] an important role is played by the phonon-spectrum branch containing a term of fourth-order in the wave vector k. In the intermediate region the dispersion law is

$$\omega^2 = ak^4 + bk_z^2, \quad k^2 = k_x^2 + k_y^2,$$

and the heat capacity of the lattice varies like T^2.

For the magnetic heat capacity of $NiCl_2$ above 14°K we used the approximation of a two-dimensional ferromagnetic system with low anisotropy[5] ($h\omega \approx 2Ask^2a_0^2$, where a_0 is the distance between the Ni ions in the layer and s is the spin, which is equal to unity for Ni^{2+})

$$C_{\text{mag}} = R\frac{\pi}{12}\frac{k_B T}{2A} = 0.0098T$$

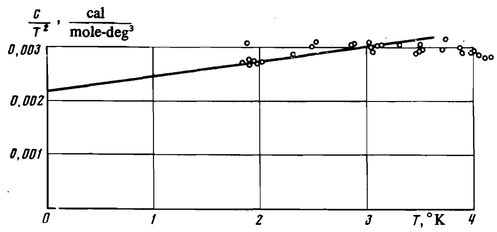

FIG. 2

and estimated the ferromagnetic interaction in the layer: $2A/k_B = 3J_1/2k_B = 53°K$, whence $J_1/k_B = 35°K$.

When separating the magnetic heat capacity of $NiCl_2$ at helium temperatures, it was assumed that the heat capacity of the lattice varies like T^3, as is the case for a number of isomorphic halides.[6,8] Figure 2 shows the temperature dependence of the heat capacity of $NiCl_2$ between 1.8 and 4°K in the coordinates C/T^2 and T. Below 3.2°K, the heat capacity can be described by the relation

$$C[\text{cal/mole-deg}]=2.8\cdot 10^{-4}T^3+2.18\cdot 10^{-3}T^2$$

where the first term can be connected with the lattice contribution, and the quadratic term with the magnetic heat capacity.

Assuming that the important terms in the dispersion of the spin waves[5] in this region are

$$(\hbar\omega)^2=(2A)^2k^4+(2B)^2k_z^2,\quad 2A={}^3/_2J_1,\quad B=J_2z_2$$

($z_1 = z_2 = 6$ is the number of nearest neighbors in the layer and between the layers, while J_1 and J_2 are the exchange integrals in the layer and between the layers), we have for the magnetic heat capacity

$$C_{\text{mag}}=R\frac{9}{4\pi^3}\frac{k_B^2T^2}{AB}=2.18\cdot 10^{-3}T^2$$

(the calculation of C for a similar dispersion law was carried out in [7]).

These low-temperature data yielded the estimate $J_1/k_B = 20°K$, it being assumed that $B/k_B = 4.6°K$; the last quantity was obtained from the value of the critical field that destroys the antiferromagnetism, $H_c = 2H_E + H_A = 129$ kOe,[9] and from the value of the high-frequency gap $\Delta_2 = (2H_EH_A)^{1/2} = 27.7$ kOe:[10,11] $H_A = 5.6$ kOe. The value $J_1/k_B = 20°K$ is approximately 30% lower than the estimate obtained by us from the linear law (above 14°K). The discrepancy is possibly due to the limited character of the employed approximations (no account is taken of the interaction of the spin waves).

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