Scientific session of the Division of General Physics and Astronomy of the Academy of Sciences of the USSR (30 October 1991)

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A scientific session of the Division of General Physics and Astronomy of the Academy of Sciences of the USSR was held on 30 October 1991 at the P. L. Kapitsa Institute of Physics Problems. The following reports were presented at the session:

1. *V. I. Nikitenko.* Topologically stable defects of the ordered state: microscopic mechanisms of generation and nonlinear dynamics.

2. *I. A. Zaliznyak, O. A. Petrenko, and L. A. Prozorova.* Properties of quasi-one-dimensional antiferromagnets with noncollinear structure.

A brief summary of one report is given below.

I. A. Zaliznyak, O. A. Petrenko, and L. A. Prozorova. *Properties of quasi-one-dimensional antiferromagnets with noncollinear structure.* Many investigators studying magnetism are now interested in antiferromagnetic dielectrics ABX_3 (A—large univalent cation, B—ion of a 3d-metal, X—halogen) with crystal structure of the type $CsNiCl_2$. The great interest in the magnetic properties of these substances is associated with two characteristics of their magnetic structure: quasi-one-dimensionality—the exchange interaction along the axis of the crystal is much stronger than the exchange interaction in the basal plane—and establishment of a noncollinear configuration of magnetic moments at temperatures below T_N (the temperature of three-dimensional magnetic ordering). Theoretical analysis shows that if only the exchange interaction of nearest neighbors is taken into account, then, because of the hexagonal symmetry, a two-dimensional triangular magnetic structure should arise: All spins are coplanar and oriented in a manner so that the angle between neighboring spins in the basal plane is close to 120° while neighboring spins along the hexagonal axis are oriented antiparallel. The relativistic interactions determine the orientation of the spins relative to the crystallographic axes. Depending on the sign of the anisotropy constant D, the plane of spin triangles can be parallel ($D>0$) or perpendicular ($D<0$) to the basal plane of the crystal. The anisotropy can also cause some distortion of the triangles.

The existence of triangular ordering has been established with the help of neutron diffraction for many ABX_3 compounds, including the easy-axis compounds $CsNiCl_3$, $RbNiCl_3$ and $CsMnI_3$ and the easy-plane compounds $CsMnBr_3$, $CsVCl_3$, $CsVBr_3$ and $CsVI_3$.

It is natural that the magnetic properties and spectrum of elementary excitations for compounds in these two groups should be substantially different. In the present work the results of electron paramagnetic resonance (EPR) investigations in a wide range of frequencies, fields, and temperatures were presented. These investigations were performed in order to determine the adequacy of the theoretical description of the real situation, to determine the phenomenological and microscopic parameters, and to study magnetic phase transitions in easy-axis ($CsNiCl_3$, $RbNiCl_3$) and easy-plane ($CsMnBr_3$) antiferromagnets with triangular ordering.

The investigations were performed on single crystals grown by Bridgman's method. The experimental apparatus consisted of a collection of direct-amplification microwave spectrometers, covering the frequency range from 0.6 to 180 GHz. The magnetic field could be varied from 0 to 60 kOe and the temperature could be varied from 1.2 to 40 K.

Since the magnetic structure of the antiferromagnets studied is determined by six sublattices, the EPR spectrum must have six branches: three acoustic and three exchange.

1. $CsNiCl_3$, $RbNiCl_3$. The experiments showed[1,2] that the phenomenological theory[3,4] describes well the complicated fields dependence of the acoustic branches of the EPR spectrum and the evolution of this spectrum as a function of the temperature (Fig. 1), and the macroscopic parameters determined from the resonance measurements (H_S—the spin-flop-transition field and the susceptibility ratio $\chi_\parallel/\chi_\perp$) agree with the data obtained from magnetostatic investigations.[5] The calculation of the EPR spectrum on the basis of the microscopic theory was performed in Ref. 6. The theoretical dependences $\nu_i(H)$ obtained in both theories for the acoustic branches agree with one another. This makes it possible to relate the macroscopic and microscopic parameters

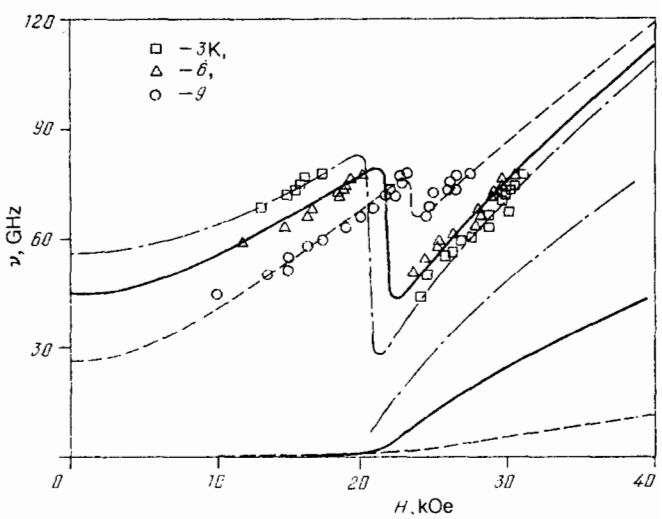

FIG. 1. Evolution of the antiferromagnetic resonance spectrum as a function of temperature in the easy-axis hexagonal antiferromagent $RbNiCl_3$ with the magnetic field oriented along the C_6 axis of the crystal.

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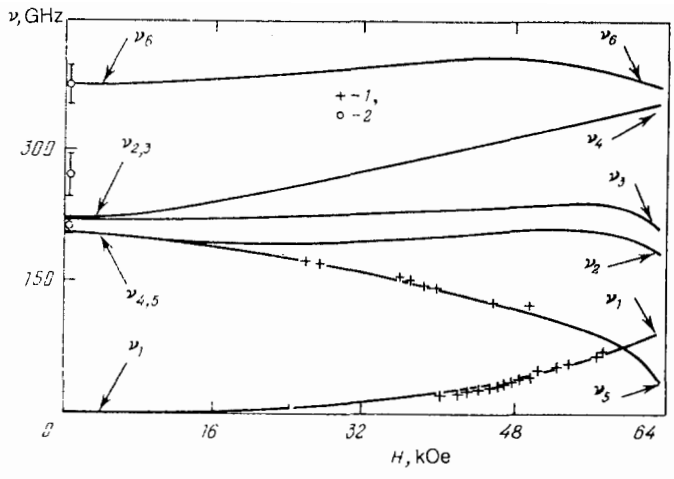

FIG. 2. AFMR spectrum in the easy-plane hexagonal antiferromagnet $CsMnBr_3$. *1*—results of our experiment at $T = 1.7$ K, $H \perp C_6$. *2*—experimental data of Ref. 14. The theoretical curves were calculated using the formulas of Ref. 12 with the following values of the constants: $\gamma = 2.8$ GHz/kOe, $J = 214$ GHz, $J' = 0.5$ GHz, and $\tilde{D} = 1.95$ GHz.

and to calculate, using the formulas from the microscopic theory, the frequencies of the exchange branches of the spectrum.

The results of such a calculation disagree significantly with inelastic neutron-scattering experiments.[7,8] This is probably due to the zero-point oscillations, which are strong owing to the quasi-one-dimensionality of these compounds, since according to Haldane's hypothesis[9,10] a system of one-dimensional antiferromagnetic chains with integer spin remains in the paramagnetic state even at zero temperature, while the excitation spectrum contains an energy gap $\nu_H \approx 0.8J$. In our case $S = 1$, but Haldane's effect is not realized in pure form, since there is a weak interaction between the spin chains and three-dimensional ordering occurs at $T \leqslant T_N$. If, however, Haldane's effect influences the EPR spectru, then it should affect primarily the exchange modes, whose frequency is comparable to ν_H. It should be noted that measurement of inelastic neutron scattering in an analogous magnetic structure in $CsMnI_3$,[11] where $S = 5/2$ and Haldane's effect is known not to occur, also does not agree with the predictions of the spin-wave theory.

2. *$CsMnBr_2$* ($D > 0$). In this substance the anisotropy is stronger than the weak in-plane exchange: $|D| > |J'|$. This results in nontrivial behavior of the spins in a magnetic field $H \perp C_6$.[12] In this case the hexagonal symmetry plays a fundamental role, and in contrast to a two-sublattice antiferromagnet, where the spins continuously follow the magnetic field, here for $D > 3J'$ spin reorientation is accompanied by a second-order phase transition: The spins remain in the plane and in the critical field $\overline{H}_c$ two pairs of sublattices collapse. As a result, the magnetic system consisting of six sublattices becomes a four-sublattice antiferromagnetic system. The existence of such a partially collapsed phase has been proved by neutron diffraction.[13] Our magnetostatic investigations have shown that the phase transition remains even when the field is taken out of the basal plane right up to the critical angle θ_c in $CsMnBr_3$ $\theta_c \approx 40°$).

Since in the case at hand the anisotropy is large, the phenomenological theory[3,4] is inapplicable, and we employed the EPR spectrum calculated on the basis of the microscopic theory (Ref. 12). According to Ref. 12, the dependences $\nu_i(H)$ are described by the roots of a system of two equations of the sixth degree. This reflects, in particular, the existence of interaction of oscillations from different branches of the spectrum, resulting in "entanglement" of the branches. The low-frequency branch itself does not have a gap at $H = 0$, and its frequency grows with the field as H^3. This unusual field dependence is associated with the fact that in the case of triangular ordering much less energy is required to turn the system of spins in a plane than in the collinear case, where $\nu \propto H$. The next mode in frequency is the exchange mode, whose energy gap at $H = 0$ is related with the critical field $\tilde{H}_c$: $\nu_5 = \sqrt{3}/2\gamma\tilde{H}_c$; at the phase-transition point the frequency of this mode becomes equal to zero.

TABLE I.

	T_N, K	H_c, $\tilde{H}_c$, kOe as $T \to 0$	J, GHz	J', GHz	D, GHz
$CsNiCl_3$	4,7 [5]	19,2	345 [16]	8,2 ± 10%	−0,59 ± 10%
$RbNiCl_3$	11 [5]	20,1	383 [17]	38 ± 10%	−1,0 ± 10%
$CsMnBr_3$	8,3 [15]	64	214 [14]	0,5 ± 5%	+1,95 ± 5%

We observed these modes experimentally (Fig. 2), and we showed that the agreement between theory and experiment is good. In the case of $CsMnBr_3$ the results also are in satisfactory agreement with neutron diffraction investigations.[14]

In conclusion we present in Table I the basic macroscopic and microscopic parameters of the investigated quasi-one-dimensional antiferromagnets with triangular ordering:

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