Magnetic resonance in the noncollinear antiferromagnet $CsNiCl_3$

I. A. Zaliznyak, V. I. Marchenko, S. V. Petrov, L. A. Prozorova, and A. V. Chubukov

Institute of Physical Problems, Academy of Sciences of the USSR; Institute of Solid State Physics, Academy of Sciences of the USSR

(Submitted 6 January 1988)
Pis'ma Zh. Eksp. Teor. Fiz. **47**, No. 3, 172–174 (10 February 1988)

The antiferromagnetic resonance spectrum during a flipping of the sublattices in the noncollinear antiferromagnet $CsNiCl_3$ has been studied experimentally and theoretically.

According to neutron diffraction studies[1] and NMR studies,[2] the compound $CsNiCl_3$ is a noncollinear antiferromagnet at temperatures below 4.4 K. The magnetic structure is determined by six sublattices of identical magnitude (Ni:S = 1). The spins of atoms which are neighbors in the hexagonal plane lie in a definite plane in spin space and are oriented at an angle of approximately 120° with respect to each other. The spins of the neighbors along the symmetry axis (the z axis), on the other hand, differ only in sign. The plane which contains the spins in the absence of a magnetic field runs perpendicular to the basal plane of the crystal, x, y. Accordingly, there are no hexagonal elements in the state of exact magnetic symmetry. Under these conditions, the fact that the angles between the sublattices are approximately 120° indicates that the structure is formed by exchange forces and that relativistic effects are small.

In order to determine the exchange symmetry,[3] it is convenient to rotate the spin space in such a way that the sublattices come to lie in a hexagonal plane (Fig. 1). It is clear from this figure that the symmetry of the particle density when relativistic effects are ignored is the same as the crystal group of the paramagnetic phase, D_{6h}^4. The exchange symmetry of the structure is specified by the antiferromagnetic vectors $\mathbf{l}_1 \perp \mathbf{l}_2$, $l_1 = l_2 = 1$, which transform under crystal transformations as the pair of functions $\cos(2\pi z/c)\cos(4\pi x/3a)$, $\cos(2\pi z/c)\sin(4\pi x/3a)$.

The anisotropy energy $\sim (v/c)^2$ in the case of this exchange symmetry reduces to

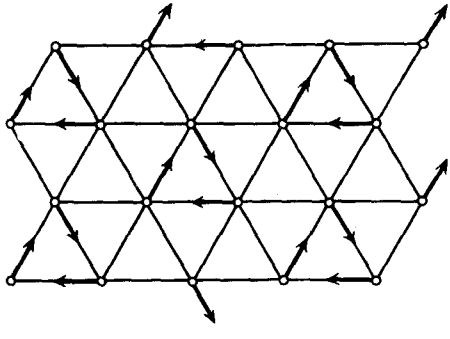

FIG. 1.

the single invariant $l_{1z}^2 + l_{2z}^2$. Introducing the unit vector $\mathbf{n} = [l_1 \cdot l_2]$, we can write the anisotropy energy in the form $\beta_z^2/2$. In $CsNiCl_3$ we have $\beta > 0$. This leading term alone is sufficient for explaining the antiferromagnetic-resonance spectrum over the frequency range which was covered in the present study.

By virtue of the exchange symmetry of the structure, the susceptibility tensor is specified by the two components $\chi_\parallel (\parallel \mathbf{n})$ and $\chi_\perp (\perp \mathbf{n})$. If $\chi_\parallel > \chi_\perp$, we would have $H = H_0 \| z$, $H_0^2 = \beta(\chi_\parallel - \chi_\perp)$, in an external field, and the sublattices should flip to a state with $\mathbf{n} \| z$. For the spectrum of the antiferromagnetic resonance in the case $H \| z$ we find[3,4]

$$H < H_0: \frac{\nu_1}{\gamma} = (\eta H_0^2 + H^2)^{1/2}, \quad \nu_2 = \nu_3 = 0;$$

$$H > H_0: \frac{\nu_{1,2}}{\gamma} = \left\{ \left(\frac{1+\eta}{2} H \right)^2 - \eta H_0^2 \right\}^{1/2} \pm \frac{1-\eta}{2} H, \quad \nu_3 = 0,$$

where $\eta = (\chi_\parallel - \chi_\perp)/\chi_\perp$ (cf. Refs. 3 and 4). The zero frequencies stem from the degeneracy of the anisotropy energy $\sim (v/c)^2$; we would have $\nu_3 = 0$ even for an arbitrary field orientation. If the external field is directed at an angle φ from the z axis, the antiferromagnetic-resonance frequencies are determined by the equation

$$\left(\frac{\nu}{\gamma}\right)^4 - \left(\frac{\nu}{\gamma}\right)^2 \{ H^2 + \eta^2 H^2 \cos^2(\theta - \varphi) - \eta H_0^2 (3\cos^2\theta - 1) + \eta \{ H_0^2 \cos^2\theta - H^2 \cos^2(\theta - \varphi) \} \times \{ \eta H_0^2 \cos 2\theta - \eta H^2 \cos^2(\theta - \varphi) - H^2 \sin^2(\theta - \varphi) \} = 0,$$

where θ is the angle between the vector $\mathbf{n}$ and the z axis, given by

$$\tan 2\theta = \frac{H^2 \sin 2\varphi}{H^2 \cos 2\varphi - H_0^2}.$$

The $CsNiCl_3$ single crystals studied in the present experiments were grown by the Bridgman method. The crystals were transparent, dark-red crystals, which cleaved well in the binary plane. The samples were parallelepipeds with linear dimensions ~ 1 mm.

The resonant absorption was measured in a direct-amplification spectrometer over the frequency range 20–80 GHz, in fields up to 35 kOe, at temperatures over the interval 1.8–46 K, and for various orientations of the crystal. The sample was held in either a short-circuited waveguide or a cylindrical resonator. The external magnetic field was calibrated on the basis of the ESR signal from DPPH, held near the sample.

In the paramagnetic phase we observe a single line $\nu = \gamma H$. At $T = 5.1$ K, we find the value $\gamma = 3.0$ GHz/kOe, in agreement with the results of Ref. 5. The linewidth is $\Delta H \sim 1$ kOe.

In the ordered phase we studied the dependence of the antiferromagnetic-reso-

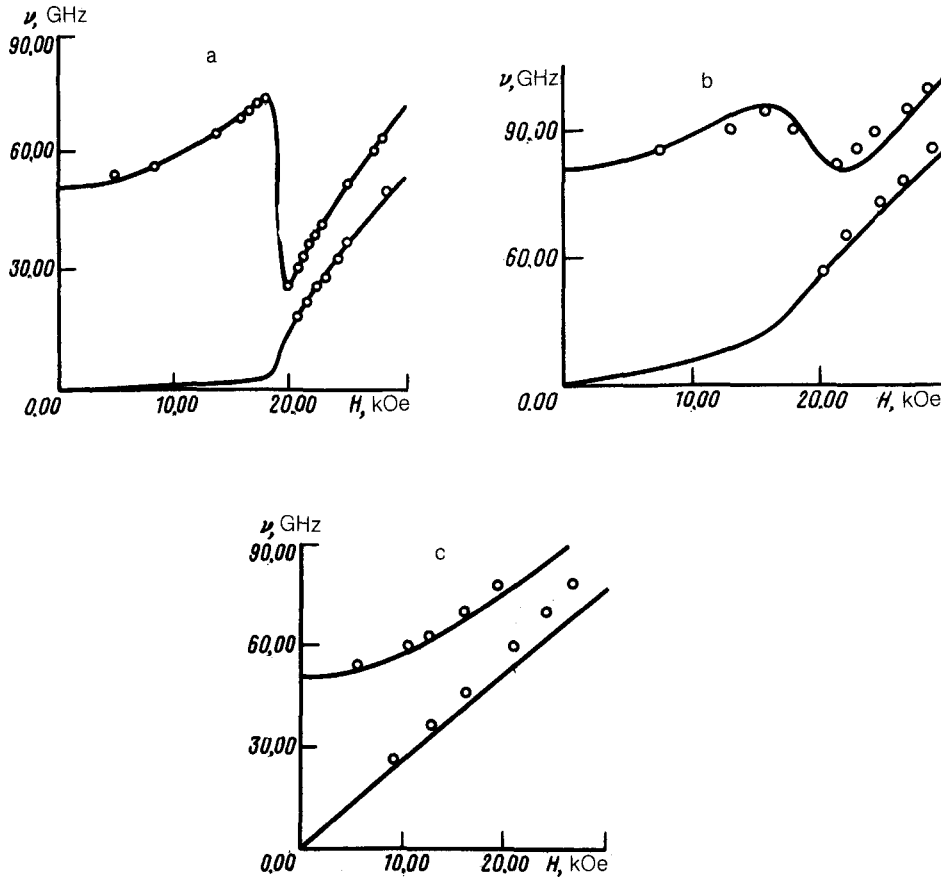

FIG. 2.

nance spectrum on the direction of the external field. Figure 2 shows the results obtained at $T = 1.8$ K (parts *a-c*, respectively, correspond to $\varphi \lesssim 1°$, $\varphi \approx 10°$, and $\varphi \approx 90°$). In fields up to the flipping point (with $\varphi \approx 0$), the line is broad, with $\Delta H \sim 4$ kOe; after the flipping we find $\Delta H \sim 0.8$ kOe. Most of the changes in the spectrum, which occurred with increasing angle φ, occurred at the small values $\varphi \lesssim 15°$. As the temperature was raised to 2.8 K, the resonance lines became markedly broader and decreased in amplitude. The frequency of the antiferromagnetic resonance at $H = 0$ decreased by 10 GHz. We plotted theoretical curves for (a) $\varphi = 1°$, (b) 10°, and (c) 90° with $\gamma = 3$ GHz/kOe, $H_0 = 19$ kOe, and $\eta = 0.8$. The results agree satisfactorily with the results of measurements of $\chi_\parallel$, $\chi_\perp$ (Ref. 5), and H_0 (Ref. 6).

According to a microscopic calculation based on the Heisenberg model, in which the interaction of nearest neighbors along the z axis (J) and in the basal plane (J') is taken into account η is approximately unity at $T = 0$ because of the small value $J' \ll J$.

[1]V. J. Minkiewicz, D. E. Cox, and G. Shirane, Solid State Commun. **8**, 1001 (1970).
[2]R. H. Clark and W. G. Moulton, Phys. Rev. **B5**, 788 (1972).
[3]A. F. Andreev and V. I. Marchenko, Usp. Fiz. Nauk **130**, 39 (1980) [Sov. Phys. Usp. **23**, 21 (1980)].
[4]L. A. Prozorova, V. I. Marchenko, and Yu. V. Krasnyak, Pis'ma Zh. Eksp. Teor. Fiz. **41**, 522 (1985) [JETP Lett. **41**, 637 (1985)].
[5]N. Achiwa, Jpn. J. Phys. Soc. **27**, 561 (1969).
[6]P. B. Johnson, J. A. Rayne, and S. A. Friedberg, J. Appl. Phys. **50**, 1853 (1979).

Translated by Dave Parsons