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LETTER TO THE EDITOR

Relativistic AFMR modes in the hexagonal antiferromagnet $CsNiCl_3$

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Abstract. The antiferromagnetic resonance spectra in hexagonal AFM $CsNiCl_3$ has been investigated both experimentally and theoretically, with special attention to relativistic modes. The very-low-energy mode at frequencies around 1 GHz has been observed. This allowed us to specify the values of the magnetic constants.

$CsNiCl_3$ belongs to a large family of hexagonal $ABCl_3$-type compounds (A = Cs, Rb; B = Ni, Co, Fe) with unusual low-temperature magnetic properties which has, in the last few years, provoked increased experimental and theoretical activity. These compounds are examples of antiferromagnets on a stacked triangular lattice with antiferromagnetic exchange interaction both in the basal XY plane (J') and along the hexagonal Z axis (J) which results in non-collinear spin ordering. Because the separation between the nearest Ni atoms in the Z direction is appreciably greater than in the XY plane and the Ni–Ni interaction in the basal plane is not direct, these compounds also demonstrate a profound quasi-one-dimensionality, that is $J \gg J'$. This was pointed out in the very first work on this topic undertaken by Achiwa [1].

For $CsNiCl_3$ it is well established by NMR and neutron diffraction experiments [2, 3] that below the $T_n = 4.4$ K Ni^{2+} ions ($S = 1$), which are neighbours in the hexagonal plane, form an approximately 120° configuration, while those along the Z axis are antiparallel. The fact that the experimentally determined magnetic structure nearly coincides with that formed by pure antiferromagnetic exchange (i.e. with a helix with the propagation vector $Q = [\frac{1}{3}\frac{1}{3}1]$) indicates that the relativistic effects in this compound are small. Nevertheless, the latter are responsible for the orientation of the spins with respect to the crystal axes. According to [2, 3], the anisotropic interactions in $CsNiCl_3$ tend to align all spins along the hexagonal axis. This, together with the non-collinearity of the magnetic structure, causes the actual spin arrangement: one of the spins in each triad is oriented along the hexagonal Z axis, while the other two are canted at an angle close to 60°. As happens in the case of the usual collinear easy-axis antiferromagnet, the application of the external magnetic field along the hexagonal Z axis causes the spin-flop transition with all the spins reorienting to be almost in the hexagonal [110] plane.

In our previous work [4] we presented experimental data on the resonant absorption in $CsNiCl_3$ measured in a direct-amplification spectrometer over the frequency range 22–80 GHz, at fields up to 35 kOe, at temperatures over the interval 1.8–46 K and for the various orientations of the crystal. Calculations, carried out in the framework of the

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macroscopic approach developed in [5], showed that when the external field is applied along the hexagonal axis **Z** and $H < H_c$, one of the relativistic frequencies grows with H as

$$\omega_1 = \gamma(\eta H_c^2 + H^2)^{1/2} \tag{1}$$

while the other two frequencies $\omega_{2,3}$, are equal to zero (i.e. the corresponding spin oscillations are the Goldstone modes). If the field exceeds the critical value H_c, one of the frequencies ω_3 remains zero while the other two almost coincide at $T \ll T_n$ and increase with the magnetic field as

$$\omega_{1,2} = \gamma\{[[(1+\eta)/2]^2 H^2 - \eta H_c^2]^{1/2} \pm [(1-\eta)/2]H\} \tag{2}$$

where $\eta = (\chi_\perp - \chi_\parallel)/\chi_\parallel$. These results are in good agreement with the experimental data [4].

The aim of the present work was to undertake a more thorough experimental and theoretical investigation of the low-energy AFMR modes in non-collinear hexagonal antiferromagnets taking $CsNiCl_3$ as an example, and to verify on this basis the numerical values of the weak exchange in the basal plane J' and the anisotropy constant D. It is worth mentioning that, in spite of the outward simplicity of the magnetic structure, up to now only the exchange integral J along the hexagonal axis has been deduced with sufficient accuracy from the available experimental data (namely, from the inelastic neutron scattering measurements near the centre of the Brilloun zone [6]): $J = 345$ GHz. The available data for J' and D differ by more than an order of magnitude [6–8].

We begin with the usual microscopic Hamiltonian, given by

$$\mathcal{H} = J\sum_{l,\Delta_z} S_l S_{l+\Delta_z} + J'\sum_{l,\Delta_\perp} S_l S_{l+\Delta_\perp} - D\sum_l (S_l^z)^2 - \gamma H \sum_l S_l^z. \tag{3}$$

Here both exchange integrals J and J' and the anisotropy constant D are positive; the exchange along the hexagonal axis is about 50 times stronger than in the basal plane, but the latter, in turn, is significantly stronger than the anisotropy.

Calculations were performed in a usual way in the framework of the six-sublattice model and the results coincided on the whole with those of the macroscopic calculations [4], which had allowed us to express the macroscopic parameters in (1)–(2) at $T = 0$ in terms of microscopic magnetic constants

$$\begin{aligned} H_c^2 &= 16\tilde{D}JS^2(1 + \tfrac{9}{4}J'/J)(1 + \tfrac{9}{8}J'/J) \\ \eta &= (1 + \tfrac{9}{4}J'/J)^{-1} \qquad \tilde{D} = D(1 - 1/2S). \end{aligned} \tag{4}$$

Besides that, the accurate account of the relativistic effects at $H < H_c$ leads to the non-zero value of the AFMR frequency ω_2 associated with the rotation of the spins in the ZX plane. Nevertheless, because of the proximity of the magnetic structure to the 120° configuration, this frequency

$$\omega_2(H) = (\omega_1(0)/2\sqrt{3})(\tilde{D}/J')[1 - (H/H_c)^2]^{1/2} \tag{5}$$

contains an additional (as compared with ω_1) small factor $\tilde{D}/J'$ and therefore was treated as being equal to zero in the macroscopic description. The possibility of observing this mode is connected with the quasi-one-dimensionality, i.e. with the exceptional smallness of the exchange integral in the basal plane (110). The third frequency, ω_3, appears also to be zero in the microscopic calculation that, naturally, reflects the invariance with respect to rotations about the Z axis.

The difficulty with the experimental observation of the AFMR mode with frequency ω_2 is associated with the necessity of having a precise orientation of the magnetic field

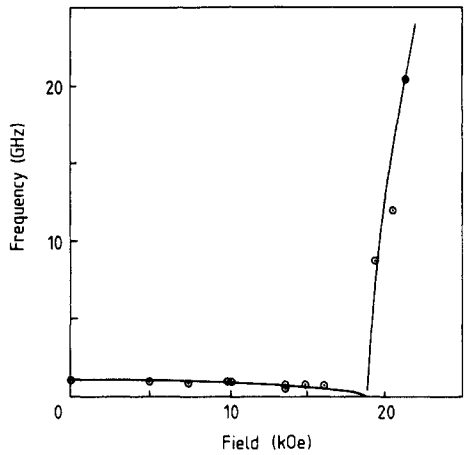

Figure 1. The AFMR spectrum in $CsNiCl_3$ at $T = 1.8$ K, $\boldsymbol{H} \| \boldsymbol{Z}$.

along the hexagonal axis, because the divergence between H and Z, even up to one, makes this mode almost dispersionless and even rising at $H \rightarrow H_c$ instead of falling [4]. In our experiments we observed the immmensely wide absorption line with $H \simeq 10$ kOe, most clearly seen at 0.8 GHz. After the frequency had been reduced to 0.6 GHz, the line disappeared. With increase in frequency it successively shifted to lower fields and ultimately disappeared at frequencies of about 1.2 GHz. Taking into account the enormous linewidth of the absorption line observed, it is quite reasonable to identify it with the weakly dispersive AFMR mode $\omega_2(H)$, see figure 1. The value of $\omega_2(0)$ obtained from our experimental data is 1.1 GHz.

Knowledge of the two relativistic AFMR frequencies at $H = 0$, $\omega_1 = 53$ GHz [4] and $\omega_2 = 1.1$ GHz, enables us to calculate the values of the basal-plane exchange integral and the anisotropy constant using the only external parameter J, known with a high accuracy from the neutron scattering study [6]. The results are: $J' = 8.2 \pm 1$ GHz; $2\tilde{D} = D = 1.18$ GHz.

It should be mentioned that the parameters discussed are connected with the long-wavelength dynamics and, hence, could differ appreciably from the actual parameters of the microscopic Hamiltonian due to zero-point oscillations. However, the quantum renormalisations do not change the functional dependencies of the relativistic frequencies [4, 5]: the expressions (4) and (5) both contain the same renormalised values of J, J' and D that we obtained by fitting to the experimental data.

In the neutron scattering study performed by Buyers and co-workers [6], the value of J' has been determined from the frequency of the highest exchange gap in the magnon spectrum. According to theory, the frequencies of the exchange modes at $H = 0$ are the following:

$$\omega_{4,5} = 6\sqrt{2}(JJ')^{1/2} \qquad \omega_6 = 12(JJ')^{1/2}. \tag{6}$$

Substituting our estimate for J' in the expression for ω_6, we obtain $\omega_6 = 638(1 \pm 0.1)$ GHz, in approximate agreement (within the accuracy limits) with the measured value $\omega_6 = 540$ GHz. In the experiments of Buyers and co-workers another resonance mode with the frequency $\omega_0 \simeq 220$ GHz was also observed. An attempt to interpret this mode as being the relativistic one leads to the enormously large value of the anisotropy constant, $D = 13$ GHz. Given such values for the constant of the Hamiltonian, the AFMR mode with the frequency 53 GHz observed in our previous experiment [4] could have corresponded to the ω_2 mode, but the excellent agreement of its experimentally

measured frequency-field dependency with that calculated for the ω_1 mode, overthrew these suggestions and unambiguously points to the mode detected at about 220 GHz being one of the exchange modes $\omega_{4,5}$. The substantial reduction of the resonant frequency measured as compared with the calculated value $\omega_{4,5} = \omega_6/\sqrt{2}$ is not clear, and needs separate, detailed consideration. In our opinion, this discrepancy could be attributed to the effect of zero-point oscillations that are particularly large for quasi-one-dimensional substances. According to Haldane [9], a one-dimensional antiferromagnet with integer S remains in the paramagnetic state even at $T = 0$. $CsNiCl_3$ does not exhibit the pure Haldane effect, since it has a finite T_n, but, because the estimated gap immediately above the singlet ground state in 1D-antiferromagnet $\omega = 0.8J = 280$ GHz [10] is of the same order as the 'classical' frequencies of the exchange modes $\omega_{4,5}$, we can expect zero-point oscillations to suppress these exchange frequencies appreciably.†

In a recent work Tanaka and co-workers [8] reported on the experimental evidence of the ω_2-like AFMR mode in $CsNiCl_3$ at frequencies of about 10 and 20 GHz. For the theoretical interpretation of the resonant absorption observed the authors of [8] supposed the unusually small value $J' = 0.6$ GHz (with $D = 1.37$ GHz), that presumes a substantial deviation of the magnetic structure from the 120° configuration (according to the classical theory the angle between the neighbours in the basic [110] plane is $\varphi = \pi - \cos^{-1}[2(1 - \tilde{D}/6J')]^{-1}$). It should be mentioned that such assumption does not agree with the results of the experimental examination of the magnetic structure [2], which yields $\varphi = 119°$. To clarify this situation, we have undertaken the additional experimental investigations of the resonant absorption in $CsNiCl_3$ at the temperature 1.8 K and $\boldsymbol{H} \| \boldsymbol{Z}$. The accuracy of the orientation was about one. A large single crystal of $CsNiCl_3$ about $2 \times 2 \times 5$ mm was either stuck at the bottom of the rectangular cavity with a Q-factor of about 2000 (for frequencies in the ranges 9–11 and 18–20 GHz) or placed inside the helix resonator (for frequencies in the range 0.5–1.5 GHz). The adequacy of the crystal structure was established with the help of powder x-ray diffraction. We also performed magnetostatic measurements with the help of a vibrating-sample magnetometer and the results obtained coincide with those reported previously [6]. In the resonance investigations at frequencies of 9, 12 and 20 GHz the single absorption line with $\Delta H \simeq 2$ kOe, located above H_c, was identified. This agrees with the results of our previous work [4]. Hence we failed to confirm the results reported in [8]. It is possible that the authors of [8] observed some kind of weak non-linear absorption, connected, for example, with the magnetoelastic interactions, which we cannot detect within our experimental accuracy of approximately 5%.

When this work was finished we learned about the work of Affleck [11], who presented a possible explanation of the 'mystery' of the $\omega_0 \simeq 220$ GHz mode. According to [11], this mode is connected not with the transverse (spin-wave-like) fluctuations, but with the longitudinal ones. This very tempting proposal needs, however, a further experimental verification.

In conclusion, we want to thank Dr Tanaka for giving us the opportunity to acquaint ourselves with the results of [8] prior to publication.

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