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Inelastic polarized neutron scattering from $S = 1$ antiferromagnetic $CsNiCl_3$ in an applied field

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Abstract. We report inelastic polarized neutron scattering measurements of the excitations in the $S = 1$, quasi-one-dimensional antiferromagnet $CsNiCl_3$ with applied magnetic field. A field up to 5.9 T was applied perpendicular to the chain direction and the field dependence of spin excitation modes at wave vectors corresponding to the one-dimensional zone centre (i.e. $(\xi, \xi, 1)$) was studied. Near the antiferromagnetic ordering point $Q = (\frac{1}{3}, \frac{1}{3}, 1)$ two types of field dependence are observed. The behaviour of the higher energy mode suggests that it is the novel longitudinal mode recently proposed by Affleck. The application of the field also reveals a near-degeneracy of the excitations observed at $Q = (0, 0, 1)$ in zero field. This degeneracy is incompatible with the result of the classical spin wave theory based on the Heisenberg Hamiltonian with a single-site anisotropy.

1. Introduction

In the past two decades the study of one-dimensional (1D) magnetic systems has proved very fruitful and has opened up new aspects in statistical physics both theoretically and experimentally [1]. These strongly fluctuating systems show a variety of non-linear excitations. Among them are topological excitations (soliton), multimagnon processes and coupled energy–spin density modes. Also, the quantum nature of 1D antiferromagnetic (AF) systems has been of great interest. After the rigorous results on the ground-state energy of $S = \frac{1}{2}$ AF chain by Bethe [2], des Cloizeaux and Pearson [3] calculated the energy of the lowest excited state. This result was then experimentally verified by Endoh *et al* [4]. The qualitative resemblance of the energy of the lowest excited state in the $S = \frac{1}{2}$ AF chain and the classical spin-wave (SW) energy dispersion based on the Néel ordered state (mis)led to the often used classical picture even for small S. There was, therefore, much controversy when in 1983 Haldane [5], using a field theoretical argument, pointed out the different nature of integer and half-integer spin value AF systems. It was especially controversial because the extreme quantum nature of the spin value dependence was derived by taking the limit of S to infinity. Among other phenomena, Haldane predicted a finite energy gap for the integer spin value 1D isotropic Heisenberg antiferromagnet (HAF) in contrast to the conventional classical SW picture. Subsequently the theoretical controversy seems to have been settled in favour of the existence of the so called 'Haldane gap' [6]. The study of the exactly solvable

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model [7], quantum Monte Carlo calculation [8] and the finite size scaling [9] supports the existence of a finite gap for the $S = 1$ isotropic HAF system. A search for a physical understanding of the difference between the half-integer and integer case has also been initiated [6, 10, 11] invoking the 'zero-spin-defect' picture.

Experimental evidence for the Haldane gap has been sought intensively by inelastic neutron scattering, because this is the only method of investigating the magnetic excitations over the whole Brillouin zone. So far three systems have been studied, namely NENP [12], $AgVP_2S_6$ [13] and $CsNiCl_3$ [14–16].

In the organic, $S = 1$ AF chain system NENP [12], Ni ions are the magnetic ions and the system does not order three dimensionally down to 1.2 K. Single-crystal inelastic neutron scattering revealed the existence of two spin fluctuation modes with energy gaps at the antiferromagnetic zone centre. They can be explained by the Haldane gap combined with a relatively large planar anisotropy. An experiment with an applied field showed only a weak field dependence of the observed gap energies for these modes [17]. On the other hand, in the recent magnetization measurements critical fields in all the crystallographic directions have been observed [18, 19], indicating the closing of the Haldane gap between the singlet and triplet states. A time of flight inelastic neutron scattering experiment on the powder sample of $AgVP_2S_6$ [13] has also indicated an energy gap. But because of the powder averaging the information available is rather limited.

$CsNiCl_3$ was studied extensively prior to the advent of the Haldane conjecture. Many different experimental methods have been applied to characterize the triangular antiferromagnetism in this quasi-1D, $S = 1$ hexagonal system [20–25], and it is well established as a quasi-1D Heisenberg system with an AF exchange and a weak axial anisotropy. $CsNiCl_3$ orders three dimensionally as a result of the interchain exchange interaction. In the ordered state, whose spin structure was known from a neutron diffraction experiment [22], the determination of the spin interaction parameters using SW theory was considered straightforward. Using unpolarized neutron inelastic scattering, Buyers *et al* [14] studied the SW excitations in the three-dimensionally ordered state and determined the microscopic parameters by applying a classical SW theory. Satisfactory agreement between the observed and calculated SW energies all over the Brillouin zone was achieved. Buyers *et al* also studied the excitations in the paramagnetic phase where 1D fluctuations dominate. The difference between the observed energy gap in the 1D phase and the gap calculated from the microscopic parameters was then interpreted as evidence for the Haldane gap. However, as pointed out by Steiner *et al* [15] on the basis of polarized neutron scattering, the agreement between observation and the classical calculation is fortuitous and the parameters obtained from this theory are inconsistent with other results. In the interpretation of the unpolarized neutron scattering results, an incorrect assignment of the higher energy mode near $(\frac{1}{3}, \frac{1}{3}, 1)$ was made, resulting in an easy axis anisotropy D which was much too large. This value of D was inconsistent with other experimental findings such as the low, spin-flop field [23] and the canting angle Θ close to 60° [22] in the ordered phase. A correct assignment of the higher energy mode near $(\frac{1}{3}, \frac{1}{3}, 1)$ results in a value of D consistent with the other observations, but causes a discrepancy between the SW calculation and the observed energy at $Q = (0, 0, 1)$ [16]. Recently Affleck [26] presented a possible explanation of this inconsistency based on the Ginzburg–Landau theory, which leads to an agreement between calculation and experiment both around $(\frac{1}{3}, \frac{1}{3}, 1)$ and $(0, 0, 1)$. One should note that in this theory the Haldane gap has been incorporated phenomenologically. Affleck explains the polarization of this higher energy mode at $(\frac{1}{3}, \frac{1}{3}, 1)$ by longitudinal spin

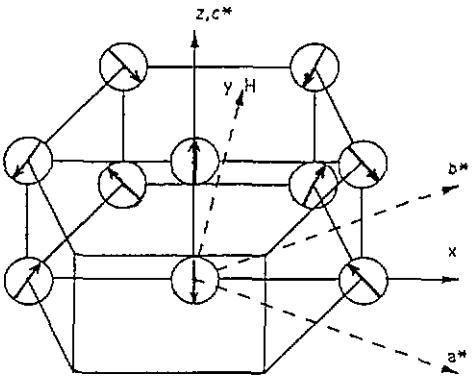

Figure 1. 3D ordered spin structure of $CsNiCl_3$ for $T < T_{N2}$ and the notation of the axes. Only one of the three possible domains is shown. The chain direction is along the $c(z)$-axis.

fluctuations which are not included in the conventional transverse SW picture. It is important to note that in the framework of the SW theory and the recent theory by Affleck one expects more than one observable mode at $\boldsymbol{Q} = (0, 0, 1)$, because x- or x, z-fluctuations are predicted to be non-degenerate with the y-fluctuations. But so far in the neutron scattering experiments only one mode has been observed at $\boldsymbol{Q} = (0, 0, 1)$.

In this paper we report a polarized neutron, inelastic scattering experiment on $CsNiCl_3$ in the ordered state at low and high applied fields in order to obtain further experimental information about the character of the magnetic transition at (0, 0, 1) and the range of validity of a SW calculation for the spin dynamics. The results show that even in the three-dimensionally ordered state neither the classical SW theory nor the Affleck approach can fully account for the experimentally observed dispersions over the whole Brillouin zone in this quasi-1D, $S = 1$ HAF system.

2. The system

$CsNiCl_3$ has a hexagonal lattice structure, space group D_{6h}^4, with lattice constants $a = b = 7.14$ Å and $c = 5.96$ Å. There are two formula units per unit cell. The magnetic moments of the Ni^{2+} ions are strongly coupled along the c-direction by the intrachain interaction J. The larger distance between the chains in the basal plane results in a weak interchain coupling J'. The general spin Hamiltonian is given by

$$H = -J \sum_{i,j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - J' \sum_{i,j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - D \sum_i (S_i^z)^2 + g\mu_B H \sum_i S_i^y \qquad (1)$$

where the first and second sums are over nearest neighbours along the chain and in the basal plane, respectively. D is the single-site anisotropy and the last term is the Zeeman energy in an applied field H in the basal plane. Because of the small, but finite J' the system orders three dimensionally at $T_{N2} = 4.40$ K. There is an intermediate phase for $T_{N1} = 4.84\ \text{K} > T > T_{N2}$, where only the z-component of the moments orders [24, 27]. The magnetic structure below T_{N2} has been determined by neutron diffraction [22]. The ordered spins lie in a plane containing one of the hexagonal symmetry directions and the c-direction. The spins are ordered antiferromagnetically along the chain direction. On one third of the chains, the spins are aligned along the c-axis and on the other chains they are canted at an angle $\pm\Theta$ away from the c-axis (see figure 1). This angle Θ is determined to be 59° at 1.6 K. In the classical theory this canting angle is determined by the ratio of J' to D. For the isotropic case the angle Θ is 60°. In the ordered state the

Table 1. Microscopic parameters as determined by different experimental methods.

Experimental method	J/k_B (K)	J'/k_B (K)	D/k_B (K)
Susceptibility [20]	−12.1		
Optical absorption [21], neutron diffraction [22]	−13 ~ −16	−0.3 ~ −0.4	0.11 ~ 0.14
Spin flop field [23]			≤0.035
Unpolarized neutron inelastic scattering [14]	−16.62	−0.29	0.63
ESR [25]	−14	−0.028	0.073
Polarized neutron inelastic scattering [16]	−16.6	−0.1	≤0.035

system has three domains due to the sixfold symmetry in the basal plane. Earlier studies have determined the microscopic parameters as listed in table 1 using the Hamiltonian (1). The determination of these parameters from macroscopic measurements is rather indirect. Optical absorption measurements and ESR give only q-averaged or $q = 0$ information respectively. Hence inelastic neutron scattering is the most direct way to obtain the microscopic parameters from the dispersion relation measured over the whole Brillouin zone. The polarized neutron inelastic scattering results are in agreement with the other results, and show that $CsNiCl_3$ can be indeed classified as a nearly isotropic HAF chain system with very small D. This contradicts the interpretation of unpolarized neutron scattering results using an incorrect assignment of spin fluctuation modes near $Q = (\frac{1}{3}, \frac{1}{3}, 1)$ [14].

3. Experiment

We performed polarized neutron experiments on the IN20 three-axis instrument at ILL. This spectrometer is equipped with Heusler-alloy monochromator and analyser crystals which polarize the neutron beams before and after scattering from the sample [28]. The horizontal collimations were chosen as 30′–40′–40′–40′ (FWHM). Energy scans in the constant Q-mode with the outgoing neutron energy fixed at $E_f = 14.7$ meV were performed. A pyrolytic graphite filter was installed behind the sample to suppress the second-order contamination. The $CsNiCl_3$ single crystal with ~1 cm^3 volume was oriented with its [110] and [001] axes in the scattering plane. A superconducting cryomagnet was used to apply a field of up to $H = 5.9$ T perpendicular to the scattering plane. A small field of 0.8 T was applied for the low field measurements to obtain a single domain state [29]. The performance of the polarized neutron beam was checked by measuring the flipping ratio which was found to be better than 19 for $H \geq 0.8$ T at a $(\frac{1}{3}, \frac{1}{3}, 1)$ AF Bragg peak.

Since the polarization direction of the neutrons along H is perpendicular to the scattering vector, the spin flip (SF) and non-spin flip (NSF) cross sections for purely magnetic scattering are [30]:

$$\begin{aligned} I_{SF} &\propto \cos^2\alpha S^{xx}(Q,\omega) + \sin^2\alpha S^{zz}(Q,\omega) \qquad \text{in SF scattering} \\ I_{NSF} &\propto S^{yy}(Q,\omega) \qquad \text{in NSF scattering} \end{aligned} \tag{2}$$

where $S^{\nu\nu}(Q,\omega)$, $\nu = x, y, z$ is the fluctuation spectrum for the ν-component of spin, and α is the angle between the scattering vector and the c-axis. At $Q = (0, 0, 1)$ a very

simple selection rule is obtained. SF corresponds to $S^{xx}(Q, \omega)$ only and NSF to $S^{yy}(Q, \omega)$ only.

4. Results and discussion

In the following, the low and high field results are presented and discussed separately.

4.1. Low field results

Figure 2 displays the measured dispersion branches in the SF and NSF channels in (*a*) the [001] and (*b*) [110] directions at $T = 2$ K. The dispersion along c has been measured using unpolarized neutrons, while the branches along the [110] direction are measured by means of polarized neutron scattering. The different polarizations of spin fluctuations seen in the SF and NSF channels are given by equations (2). In the insets of figure 2 the temperature dependences of the excitation energies at (*a*) $Q = (0, 0, 1)$ and (*b*) $Q = (\frac{1}{3}, \frac{1}{3}, 1)$ are indicated. Approaching T_N from above the softening of the mode at $Q = (\frac{1}{3}, \frac{1}{3}, 1)$, the AF Bragg point for the spin structure shown in figure 1, is clearly seen. Below T_N only the higher energy mode is seen at $(\frac{1}{3}, \frac{1}{3}, 1)$. In contrast, the excitation energy at $(0, 0, 1)$ is hardly affected by the three-dimensional (3D) ordering.

In figure 3(*a*) the low field result ($H = 0.8$ T) at $Q = (0, 0, 1)$ obtained using polarized neutrons is shown. One can see in the NSF scattering a resolution limited peak around 0.5 THz that, according to equation (2), has to be associated to the y-mode. In the SF scattering a somewhat broader single peak is observed at the same energy, which must be associated with x-fluctuations according to equations (2).

4.1.1. Discussion. In the frame of the classical SW theory the set of microscopic parameters for the Hamiltonian (1) is uniquely determined by the energies at the 3D zone centre and 1D zone boundary. The intrachain exchange J is determined by the excitation energy at $(0, 0, 1.5)$. The upper limit of the single site anisotropy is given by the fact that an energy gap in the y-mode at $(\frac{1}{3}, \frac{1}{3}, 1)$ is not observed even in the high-resolution measurement (energy width ~0.1 meV FWHM) using the cold-neutron 3-axis spectrometer IN12 [15]. The polarized neutron scattering clearly shows that the polarization of the higher energy mode near $(\frac{1}{3}, \frac{1}{3}, 1)$ is x–z like. Thus the adjustment of the x–z mode to the observed mode gives the following set of parameters: $J/k_B = -16.6$ K, $J'/J = 0.006$ and $D/k_B \leqslant 0.035$ K (see table 1).

The lines in figure 2 indicate the results of the classical SW calculation using these parameters. Clearly there is a disagreement between the calculated and observed dispersion at $Q = (0, 0, 1)$. First of all, the predicted energies are not correct. Furthermore, the classical SW theory predicts x- and z-fluctuations being non-degenerate with the y-mode. The polarized neutron results shown in figures 2 and 3(*a*) unambiguously demonstrate the following.

(i) Even in the 3D ordered state, a consistent classical SW description over the whole Brillouin zone is not possible in this $S = 1$, quasi-1D HAF system.

(ii) At $Q = (0, 0, 1)$ there is not only a quantitative, but qualitative disagreement between the experiment and the classical SW prediction. Namely the y- and x-modes at $(0, 0, 1)$ are nearly degenerate within resolution of the present experiment.

Affleck recently proposed a model for a lattice of weakly coupled spin chains [26]. He maps the problem on to a lattice of coupled (1 + 1)-dimensional field theory. In this

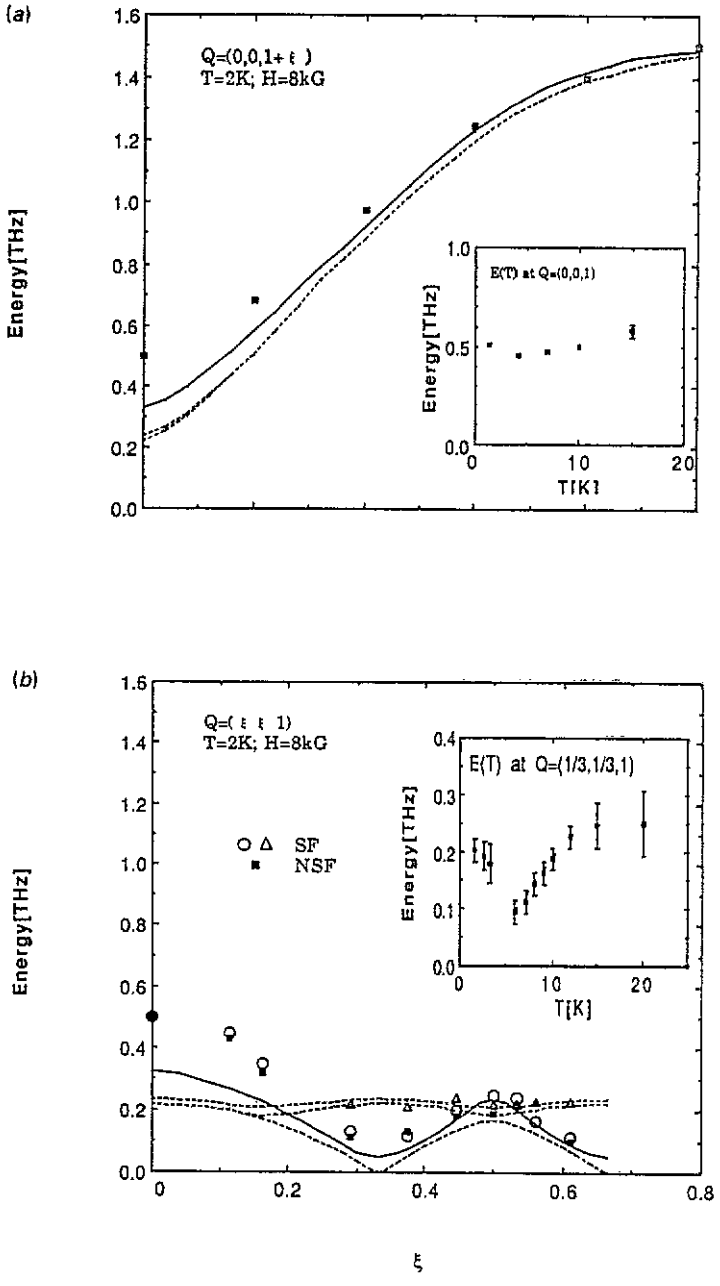

Figure 2. sw dispersion in $CsNiCl_3$ at $T = 2$ K and $H = 0.8$ T. (*a*) The dispersion in the [001] direction measured by unpolarized neutron, inelastic scatteirng. Inset: the temperature dependence of the mode at $Q = (0, 0, 1)$. (*b*) The dispersion in the [110] direction measured by polarized neutron, inelastic scattering. Inset: the temperature dependence of the mode at $Q = (\frac{1}{3}, \frac{1}{3}, 1)$ observed by unpolarized neutron, inelastic scattering. The spin wave energies resulting from the Hamiltonian (1) and the parameters described in the text are also indicated. The full and broken curves indicate y- and x, z-modes, respectively.

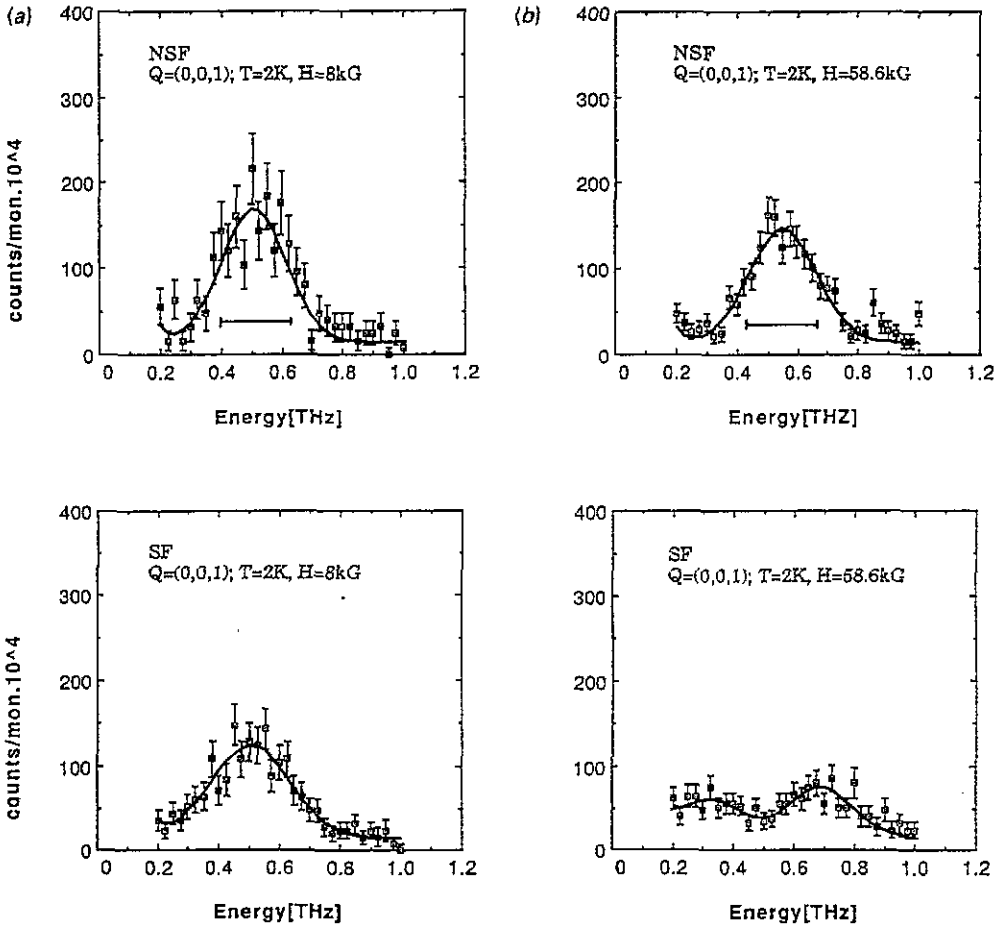

Figure 3. (*a*) NSF and SF spectra at $Q = (0, 0, 1)$ for $T = 2$ K and $H = 0.8$ T. (*b*) NSF and SF spectra at $Q = (0, 0, 1)$ for $T = 2$ K and $H = 5.9$ T. Counts are per 10^4 monitor counts, corresponding to roughly 22 min counting time. The full curves are fits with Gaussians. The horizontal bar indicates the instrumental resolution.

approach, the upper branch near $(\frac{1}{3}, \frac{1}{3}, 1)$ is a longitudinal mode, with fluctuations along the local spin directions (i.e. x, z fluctuations), and the experimental findings near $(\frac{1}{3}, \frac{1}{3}, 1)$ are reproduced with $D = 0$. But here again the degeneracy at $Q = (0, 0, 1)$ is lifted due to the J', so that in principle the x-fluctuations around 0.4 THz, i.e. just below the y-mode, should be observable by neutron scattering as predicted in the SW theory too. So the qualitative disagreement between the experimental result and theoretical prediction at $Q = (0, 0, 1)$ remains even in Affleck's model.

The temperature dependence of the mode at $Q = (0, 0, 1)$ and $(\frac{1}{3}, \frac{1}{3}, 1)$ suggests that there are regimes on the 1D zone centre plane, at which the low dimensional character is retained even below T_{N2} (in the 3D ordered state). Only around the 3D ordering points does one observe the effect of the ordering and SW theory works. We mention that a similar behaviour is observed in singlet ground state systems, where the interchain exchange is large enough to induce the 3D ordering at a finite temperature [31]. There the temperature dependence of the Γ-magnon energy (i.e. at $Q = (0, 0, 1)$) and the

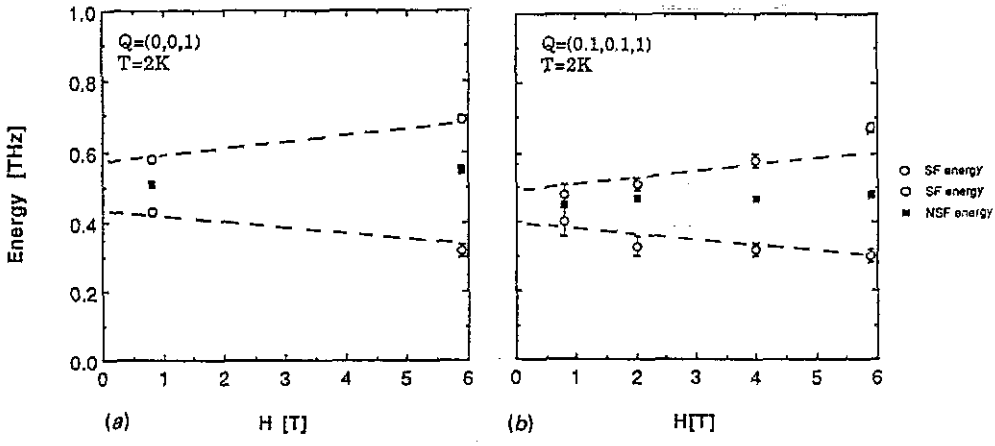

Figure 4. Field dependence of the observed excitation energies at (*a*) $Q = (0, 0, 1)$ and (*b*) $Q = (0.1, 0.1, 1)$ for $T = 2$ K. The modes observed in SF and NSF are indicated by open circles and full squares respectively. The broken lines are guides for the eye with a slope corresponding to a Zeeman splitting with $1.05\mu_B$.

softening of the K-magnon (i.e. at $Q = (\frac{1}{3}, \frac{1}{3}, 1)$) is described by the dynamical correlated effective field approximation (DCEFA) [32] for the singlet–doublet system. But in contrast to the singlet ground state system we observe quasielastic scattering here indicating 1D correlation above T_N. We measured the correlation length $\xi(T)$ [33] and obtained for example, $\xi \cong 60$ Å at $T = 7$ K and $Q = (0.52, 0.52, 1 \pm \eta)$.

The observed discrepancy between the SW calculation and the experimentally observed dispersion, in particular around (001), points to the importance of the Haldane ground state even for the 3D ordered state. This is further substantiated by the agreement, as far as the excitation energies are concerned, between experiment and Affleck's prediction which includes the Haldane gap [26]. Although the eigenvectors of the observed modes have been determined, a conclusion on the character of the ground state is not yet possible.

4.2. High field results

To obtain more insight into the ground state properties of this system, we applied a transverse field up to 5.9 T in the hope that the field dependence of the modes might yield more information on the degeneracy and on the eigenvectors, and hence on the discrepancies between the classical SW picture and experiment.

In figure 3(*b*) the spectra at $Q = (0, 0, 1)$ in the applied field of $H = 5.9$ T are presented. In the NSF spectrum the peak position is only slightly shifted with respect to the low field result, but the SF scattering now clearly indicates a two peak structure. This high field result demonstrates that the single mode observed at $Q = (0, 0, 1)$ in the unpolarized neutron experiment is indeed nearly threefold degenerate.

In figure 4 the field dependence of the observed excitation energies are shown at $Q = (0, 0, 1)$ and $(0.1, 0.1, 1)$. In both cases the splitting of the SF mode is clearly visible at the highest applied field, while the energy of the NSF mode only increases slightly. The splitting corresponds to a magnetic moment of about 1 μ_B. (The broken lines in figure 4.) The line width of the NSF scattering hardly changes with field, while the SF line width would have to be assumed to increase drastically if a single-mode picture were to be

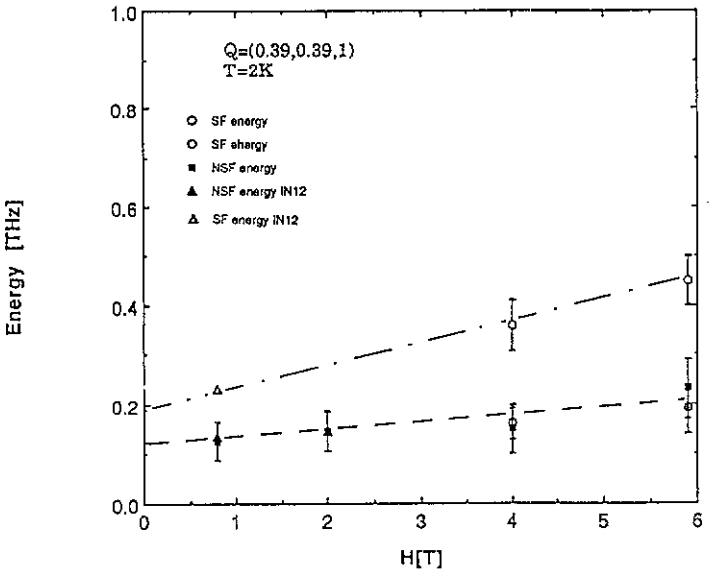

Figure 5. Field dependence of the excitation energies at $Q = (0.39, 0.39, 1)$ for $T = 2$ K. The high resolution results for low fields obtained with the IN12 spectrometer at ILL [15] are also included. The broken line has the same meaning as in figure 4. The chain line is a guide for the eye with double the slope of the broken line.

retained at low fields. The result shown in figure 4 is obtained by assuming a field independent line width for SF scattering which is comparable to the measured line width of the NSF mode.

In figure 5 the field dependence of the excitation energies at $Q = (0.39, 0.39, 1)$, near the 3D AF ordering point $(Q = (\frac{1}{3}, \frac{1}{3}, 1))$ is shown. Here the lower branch is composed of NSF and SF scattering and the upper branch of purely SF, as has been demonstrated by the previous polarized neutron scattering experiment [15]. By increasing the field, both the NSF and SF components of the lower branch display a Zeeman energy increase corresponding to a moment of around 1 μ_B (broken line in figure 5). The upper branch, however, shows an increase with the field, that is roughly twice as steep (indicated with the chain line in figure 5) as the expected Zeeman increase for the magnetic moment of $1\mu_B$. We mention that the line width of the upper branch seems to increase with the applied field, but because of the coarse energy resolution in this experiment a definite statement requires a high resolution measurement.

Thus the application of a magnetic field to $CsNiCl_3$ results in two remarkable features of the observed excitation modes.

(i) The mode at (001) is split into three, of which two (the SF modes) show a Zeeman splitting which is characterized by a magnetic moment of approximately $1\mu_B$, while the NSF mode is field independent.

(ii) All modes around $(\frac{1}{3}, \frac{1}{3}, 1)$ increase in energy with the applied field. The low energy modes increase by a Zeeman energy corresponding to a magnetic moment of $1\mu_B$. The higher energy mode shifts with the Zeeman energy corresponding to a moment of approximately $2\mu_B$.

4.2.1. Discussion. These high field results show that the number of modes at $Q = (0, 0, 1)$, namely three, predicted by the SW theory and Affleck's calculation agrees with

the experimental findings. The extrapolation to zero field indicates that the two SF modes do not really merge into a single mode at $H = 0$, so the observed intrinsic linewidth of the SF mode at $H = 0.8$ T is due to this small splitting of about 0.1 THz. This result shows that in zero field there are indeed three nearly degenerate modes at $Q = (0, 0, 1)$. Because of the non-observability of the z-fluctuations at $Q = (0, 0, 1)$, the observed mode cannot be uniquely associated with the pure x- and z-modes predicted by conventional SW theory. Affleck predicts two modes of xz character but they lie roughly 0.15 THz below the y-mode and not around the y-mode as found experimentally.

The changes with field in the energies of the SF branches are roughly comparable with the expected Zeeman splitting when assuming the Ni^{2+} moment of $1.05\mu_B$ as established by Cox and Minkiewicz [34] at low T. Because Ni^{2+} with nearly octahedral coordination in the ionic compounds is usually found to have a moment around $2\mu_B$, Cox and Minkiewicz ascribe the reduction of the moment to the strong fluctuating nature of this quasi-1D system.

We mention that in $CsFeCl_3$, a singlet ground-state ferromagnetic chain system with an excited doublet state, a Zeeman splitting of the gap mode corresponding to the excitations from singlet to doublet states is observed with applied field [35]. There the splitting corresponds to a moment of $2.5\mu_B$, expected for the Fe^{2+} ion. This result in the singlet-doublet case is explained by DCEFA [32] of such a system.

The strong reduction of the Zeeman splitting in the AF system points to the strongly fluctuating character of the AF ground state even in the 3D ordered state.

Since the mode at $Q = (0, 0, 1)$ hardly changes at the 3D phase transition, we interpret its field dependence as a property of the individual chain of this quasi-1D system with $S = 1$. In fact, recent high field measurements at elevated temperatures above T_N indicate the same Zeeman splitting at $Q = (0, 0, 1)$ [36]. Hence the fact that the mode at $Q = (0, 0, 1)$ in $CsNiCl_3$ experimentally splits into at least three different modes upon applying the field may suggest that the original mode is a triplet with an energy gap to a non-magnetic ground state. It can be speculated that this non-magnetic ground state is the new collective singlet ground state of the quantum $S = 1$ chain system conjectured by Haldane.

If one extrapolates the closing of the gap due to the splitting of the SF branch at $Q = (0, 0, 1)$, one obtains a critical field of ~18 T. One should note the difference in the field dependence of the gap mode in the NENP case where at $k = \pi$ no such closing of the gap is observed in neutron scattering [17], while magnetization measurements do indicate a closing of the gap [18, 19].

The different field behaviour of the modes near $(\frac{1}{3}, \frac{1}{3}, 1)$ (figure 5) can be understood as an indication for the novel longitudinal nature of the fluctuations associated with the higher energy mode as proposed by Affleck. He has suggested the original triplet of the single chain breaks up into a pair of Goldstone modes and a longitudinal mode. The two Goldstone modes are transverse fluctuations of the conventional SW type, while the longitudinal fluctuations are not obtained by the conventional theory: they are associated with the fluctuations along local spin directions. Affleck does not give any prediction for the field dependence of this longitudinal mode, but it is likely that this kind of longitudinal fluctuations has similarities with multimagnon processes [37, 38], and thus could show a different field dependence from that of the normal transverse fluctuations.

5. Summary

To conclude, polarized neutron inelastic scattering from the $S = 1$, quasi-1D, nearly isotropic Heisenberg antiferromagnet $CsNiCl_3$ clearly showed that neither the classical

spin wave theory nor the recent theory by Affleck fully accounts for the experimentally observed dispersion relation in the 3D ordered phase. The results presented here therefore point to the influence of the Haldane ground state, even in the 3D ordered regime, in particular around the quasi-1D Brillouin zone centre (0, 0, 1). The observed splitting at (0, 0, 1) in a magnetic field closely resembles the behaviour of a system with crystal field singlet ground state atoms. The observed reduction of the magnetic moment in the Zeeman energy can be seen as evidence for strong 1D collective fluctuations of singlet character, which are considered essential to the Haldane ground state.

It is obvious that the experimental information now available about the ground state of this quasi-1D Heisenberg antiferromagnet with $S = 1$ cannot be explained without a thorough theoretical treatment of the influence of a magnetic field on the ground state and the excitations of a Haldane system. We urge that such work be initiated.

Acknowledgments

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Note added in proof After the acceptance of our paper we were informed by I Affleck that in his original paper (see [26]) the mixing between the transverse xz and longitudinal modes was overlooked (see Erratum in 1990 *Phys. Rev. Lett.* **65** 2477). Consequently there is only one visible xz mode, but two-fold degenerate, at $Q = (0, 0, 1)$ in the ordered state. Furthermore in the limit of J' reduced to its critical value where order disappears, and neglecting the anisotropy, these xz and y modes at (0, 0, 1) become exactly degenerate to form the triplet spectrum obtained for the one-dimensional phase.

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