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Magnetic gap excitations in a one-dimensional mixed spin antiferromagnet Nd_2BaNiO_5

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Magnetic excitations in a Nd_2BaNiO_5 powder sample were investigated by inelastic neutron scattering. Well-defined one-dimensional (1D) gap modes were observed *above and below* the Néel temperature $T_N = 24$ K. In both temperature regimes the measured dynamic structure factor was found to be consistent with that calculated for transverse Haldane gap modes in a quantum-disordered $S=1$ linear-chain antiferromagnet. The energy gap does not show any appreciable T dependence at $T>T_N$. However, in the ordered phase the spin gap *increases*. The observed behavior is totally different from that found in other Haldane gap systems undergoing 3D antiferromagnetic ordering, such as $CsNiCl_3$. [S0163-1829(96)06134-6]

I. INTRODUCTION

Since the pioneering work of Haldane[1,2] much attention has been given to quantum magnetism of quasi-one-dimensional (1D) integer-spin Heisenberg antiferromagnets (AF). If the interaction between the AF chains is substantially weaker than the in-chain exchange coupling, such systems have a nonmagnetic ground state (i.e., with no long-range order in the spin pair correlation function) and an exchange-induced gap in the magnetic excitation spectrum. The low-lying magnetic excitations are singlet to triplet, and the dispersion has a minimum (referred to as the Haldane gap) at the 1D AF reciprocal space point. Fortunately, this unique, purely quantum behavior is relatively robust. Weak easy-plane single-ion anisotropy does not destroy the nonmagnetic ground state or eliminate the energy gap, although it does indeed split the excitation triplet into a lower-energy doublet of transverse modes (with spin fluctuations perpendicular to the chain direction) and a higher-energy longitudinal mode (spin fluctuations along the chains).[3–5] Moreover, the gap can be observed at finite temperatures. This robustness made it possible to study the Haldane gap experimentally in quite a number of real quasi-1D compounds, and inelastic neutron scattering proved to be the most informative technique (for a comprehensive reference list see, for example, Refs. 6–8).

In this work we focus on the isostructural family of quasi-1D materials with the general formula L_2BaNiO_5. Members with $L=$Y, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, and Tm are known.[9,10] In these species the high-spin ($S=1$) Ni^{2+} ions are arranged in linear chains along the **a** axis of the orthorhombic structure, and the in-chain antiferromagnetic exchange constant is relatively large ($|J| \approx 250$ K).[11,12] For nonmagnetic L ($L=$Y) the interchain interaction is negligible. Y_2BaNiO_5 is a good example of a Haldane-gap system[13,14,11] and does not exhibit any long-range magnetic order down to 1.2 K. The L sites being positioned in between the Ni chains, substituting magnetic ions for L leads to an increased interchain coupling and all the $L \neq$ Y members order antiferromagnetically with T_N ranging from 24 to 50 K (Ref. 15 and references therein). It is known that the onset of 3D long-range order at low temperature does not eliminate the possibility of Haldane-like behavior.[16–19,8] However, purely 1D systems appeared to be of greater immediate fundamental interest. Much of the experimental work, especially inelastic neutron scattering, was focused on Y_2BaNiO_5 and until recently the $L \neq$ Y members were largely neglected. Our studies of Pr_2BaNiO_5 (which orders at $T_N = 24$ K) have revealed interesting and unique dynamic properties of this *mixed spin* system.[12] In particular, well-defined Haldane-like gap excitations were observed at $T>T_N$ and shown to persist in the 3D AF phase. Moreover the gap mode retains its purely 1D dispersion in the ordered phase. In contrast with the behavior observed in such materials as $CsNiCl_3$,[17,16,8] no softening is observed in the gap mode at any wave vector. The gap excitations are distinct from acoustic spin waves in the ordered state. It was also demonstrated that these excitations represent spin fluctuations in the Ni-chain subsystem, with hardly any contribution from the rare-earth sites.

Herein we report the results of inelastic neutron-scattering experiments on Nd_2BaNiO_5. We show that a well-defined 1D magnetic transverse gap mode is present in Nd_2BaNiO_5 both above and below the Néel temperature $T_N = 48$ K (Ref. 15). It corresponds to the Haldane-gap excitations in Y_2BaNiO_5. Unlike those in the extensively studied $CsNiCl_3$ compound,[17,16,8] the gap modes in Nd_2BaNiO_5 do *not* experience any softening at T_N at any wave vector and are apparently *not* related to the gapless antiferromagnons (Goldstone modes) in the ordered phase. The results are discussed in comparison with those previously obtained for Pr_2BaNiO_5.

II. EXPERIMENTAL

The preparation of Nd_2BaNiO_5 samples is described elsewhere.[15] We have utilized the same single-crystal sample ($3\times2\times1$ mm^3) that was used by Sachan *et al.*,[15] for the studies of the magnetic structure and a newly synthesized 15 g powder sample. The crystal structure of Nd_2BaNiO_5 is orthorhombic (space group *Immm*), with cell constants

$a=3.832$ Å, $b=5.926$ Å, and $c=11.652$ Å at room temperature. The antiferromagnetic transition occurs at $T_N=48$ K, giving rise to magnetic Bragg reflections of type $((2m+1)/2\ k\ (2n+1)/2)$ (m,k,n integer).[15] The magnetic ordering in our powder sample was checked in a simple neutron powder-diffraction experiment.

Inelastic neutron-scattering experiments were carried out on the H4M and H8 triple-axis spectrometers at the High Flux Beam Reactor at Brookhaven National Lab. The single-crystal sample was mounted with the (0 1 0) crystallographic plane parallel to the scattering plane of the spectrometer, making $(h\,0\,l)$-type scattering vectors accessible for measurement. The use of a standard Displex refrigerator enabled us to perform the measurements in the temperature range 11–300 K. To meet the conflicting needs of resolution and intensity several spectrometer configurations were exploited. For the single-crystal experiments $40'-40'-80'-80'$ collimations and fixed final energy neutrons ($E_f=14.7$ meV) were used, with a pyrolitic graphite (PG) filter positioned after the sample. This setup yields an energy resolution of $\delta E_0\approx 1.4$ meV at $\hbar\omega=0$. Measurements on powder samples were performed using $40'-40'-40'-80'$ collimation and fixed final ($E_f=14.7$ meV, $\delta E_0\approx 1.1$ meV or $E_f=30.5$ meV, $\delta E_0\approx 3.5$ meV) or fixed incident ($E_i=30.5$ meV, $\delta E_0\approx 3.5$ meV) neutron energies, and PG filters placed in front of the analyzer or in front of the sample, respectively.

III. RESULTS

A. Single-crystal experiments

The Nd_2BaNiO_5 single crystal available for our studies is barely large enough for magnetic inelastic scattering. Figure 1 shows a constant-**Q** scan taken at the 1D AF point $\mathbf{Q}=(1.500)$ at $T=55$ K (solid circles). A broad inelastic peak is observed at $\hbar\omega\approx 11$ meV, roughly the same energy as the transverse Haldane gap in Y_2BaNiO_5. The feature is not present at $\mathbf{Q}=(1.700)$ (open circles), which is consistent with the steep dispersion along the chain direction, expected for Haldane-gap excitations.

B. Powder experiments

Single-crystal measurements are as a rule more informative than those performed on powder samples. However, often one can obtain higher quality data in a powder experiment, simply because a larger sample volume is available. One complication arises from the fact that there are several relatively low-energy crystal-field (CF) excitations associated with Nd^{3+}. These dispersionless modes produce inelastic peaks much stronger than those originating from the 1D excitations this paper is concerned with. For this reason we have carefully mapped out the CF excitations, checking their Q and T dependences, as described below in a separate section. We have identified a ‘‘clean’’ energy region, 5 meV $\leqslant\hbar\omega\leqslant 15$ meV, free of CF transitions, for studying the 1D Ni-chain modes.

1. Powder-averaged magnetic cross section

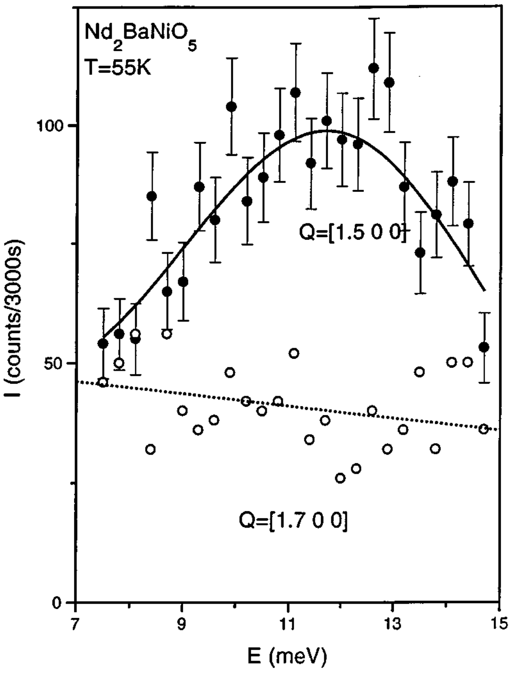

FIG. 1. A constant-**Q** scan for the Nd_2BaNiO_5 single crystal sample measured at the 1D antiferromagnetic reciprocal space point [1.500] (solid circles). The solid line is a guide to the eye (Gaussian fit) and the dotted line represents a linear fit to the background measured at [1.7 0 0] (open circles).

To properly interpret the results of inelastic neutron-scattering experiments on powder-averaged samples one has to consider the powder dynamic structure factor $S_{\rm pow}(Q,\omega)$ for a 1D Haldane gap antiferromagnet. For single-crystal samples the following empirical form of the magnetic cross section $S(\mathbf{Q},\omega)$ has been successfully implemented to analyze inelastic neutron-scattering data collected around the 1D AF planes $((2m+1)/2\ 0\ 0)$ (m integer) for several Haldane-gap systems:[7,6]

$$S_m(\mathbf{Q},\omega)=S_m(Q_\parallel,\omega)\propto\frac{|f(Q)|^2}{\sqrt{\kappa^2+(Q_\parallel-Q_m)^2}}\,\delta(\hbar\omega-\hbar\omega_{Q_\parallel}). \tag{1}$$

Here $Q_\parallel$ is the component of the scattering vector **Q** along the chain direction, κ is the inverse instantaneous spin-correlation length, and $f(Q)$ is the magnetic form factor (for Ni^{2+} in our case). We have introduced the notation $Q_m=(2m+1)(\pi/a)$, where a is the intrachain spin-spin separation (which in L_2BaNiO_5 compounds coincides with the a cell constant). In Eq. (1) $\omega_{Q_\parallel}$ denotes the 1D dispersion relation, for which the quadratic approximation is usually used:

$$\hbar\omega_{Q_\parallel}=\sqrt{\Delta^2+c_0^2(Q_\parallel-Q_m)^2}. \tag{2}$$

In this formula Δ stands for the Haldane gap and c_0 is the spin-wave velocity along the chains.

In general, for 1D excitations $S(\mathbf{Q},\omega)=S(Q_\parallel,\omega)$ and the powder-averaged dynamic structure factor is given by

$$S_{\rm pow}(Q,\omega)=\frac{1}{4}\int_{-1}^{1}S(Qz,\omega)P(z)dz, \tag{3}$$

where $P(z)$ is the polarization factor for unpolarized neutrons.[20] In a system with easy-plane single-ion anisotropy the lower Haldane doublet corresponds to fluctuations of the spin components perpendicular to the chain direction (transverse modes), and $P(z)=(1+z^2)$ (Ref. 13).

Note, that Eqs. (1) and (2) are valid only in the vicinity of the 1D AF reciprocal lattice planes. However, the dispersion along the chain direction is very steep, and for small energy transfers (less than several units of Δ) both equations adequately describe the cross section for all $Q_\parallel$.

Combining Eq. (1) with Eq. (3) and using $S(Q_\parallel,\omega)=\Sigma_m S_m(Q_\parallel,\omega)$, we find that the integration is canceled by the the δ function. Introducing the notation

$$z_m^\pm(\omega)=\frac{1}{Q}\left[Q_m\pm\frac{1}{c_0}\sqrt{(\hbar\omega)^2-\Delta^2}\right], \tag{4}$$

the following result is easily obtained:

$$S_{\text{pow}}(Q,\omega)=s(Q,\omega)F(\omega), \tag{5}$$

$$s(Q,\omega)=\frac{|f(Q)|^2}{Q}\left[\sum_{m:|z_m^+(\omega)|\leqslant 1}[1+\{z_m^+(\omega)\}^2] + \sum_{m:|z_m^-(\omega)|\leqslant 1}[1+\{z_m^-(\omega)\}^2]\right], \tag{6}$$

$$F(\omega)\propto\frac{1}{\sqrt{\kappa^2c_0^2+(\hbar\omega)^2-\Delta^2}}\times\frac{\hbar\omega}{\sqrt{(\hbar\omega)^2-\Delta^2}}. \tag{7}$$

For a Haldane-gap AF the quantities Δ, κ, and c_0 are not independent but are related by $\kappa c_0=\Delta$ (Refs. 3, 21, and 22), and Eq. (7) is much simplified:

$$F(\omega)\propto\frac{1}{\sqrt{(\hbar\omega)^2-\Delta^2}}. \tag{8}$$

Conveniently, for Q not too close to Q_m $[|Q-Q_m|>\sqrt{(\hbar\omega)^2-\Delta^2}]$, $s(Q,\omega)$ is ω independent. Indeed, $1+(z_m^\pm)^2\approx 1+(Q_m/Q)^2$ for small energy transfers, and

$$s(Q,\omega)\approx S_{\text{pow}}(Q)=\frac{|f(Q)|^2}{Q}\sum_{m:|Q_m|\leqslant Q}[1+(Q_m/Q)^2]. \tag{9}$$

In other words, if the measurements are performed away from the 1D AF wave numbers, the powder cross section can be decomposed into a product of a Q-dependent part $S_{\text{pow}}(Q)$ and a Q-independent spectral weight function $F(\omega)$.

2. Results

To maximize the magnetic signal from the powder sample constant-Q scans were performed close to the maximum of $S(Q)$ at $Q=0.65\times\pi/a$. Using a fixed incident neutron energy $E_f=14.7$ meV limits the accessible energy range to ≈ 14 meV for this wave vector. The data collected at several temperatures (both above and below T_N) are shown in Fig. 2. The gap at $\Delta\approx 11$ meV is readily observed, although the scans could not be extended to sufficiently large energies to cover the entire peak. The solid lines in Fig. 2 show fits with Eq. (8), convoluted with the experimental energy resolution [full width at half maximum (FWHM)$\approx$2.5 meV at $\hbar\omega=11$ meV]. These fits were obtained by refining the gap energy Δ, the overall scaling factor, and a flat background. Δ is plotted against temperature in Fig. 3. Below $T=30$ K the gap could not be observed within our scan range.

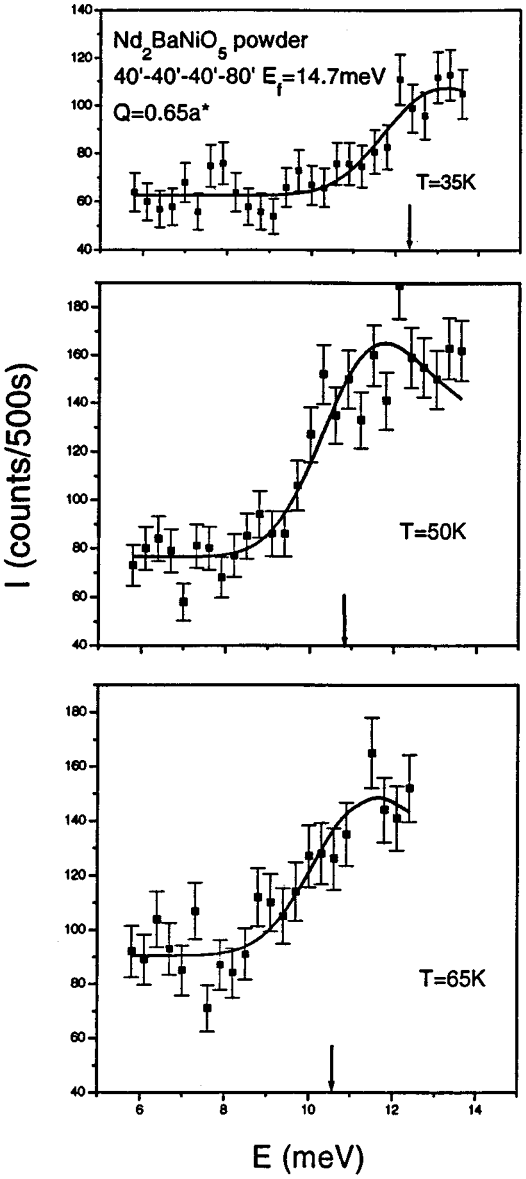

FIG. 2. Constant-Q scans measured for the Nd$_2$BaNiO$_5$ powder sample at several temperatures. The solid line represents the fits described in the text. Arrows indicate the position of the energy gap.

To verify the 1D nature of the gap excitation and to prove that it corresponds to spin fluctuations perpendicular to the Ni-chain direction we performed constant-E scans, with the spectrometer tuned to roughly the gap energy (Fig. 4, solid circles). The background, measured at $\hbar\omega=6$ meV is shown in open circles. Both *above and below* T_N the scans have a typical 1D profile with asymmetric peaks at the 1D AF wave numbers (π/a, $3\pi/a$, etc.), similar to those observed in

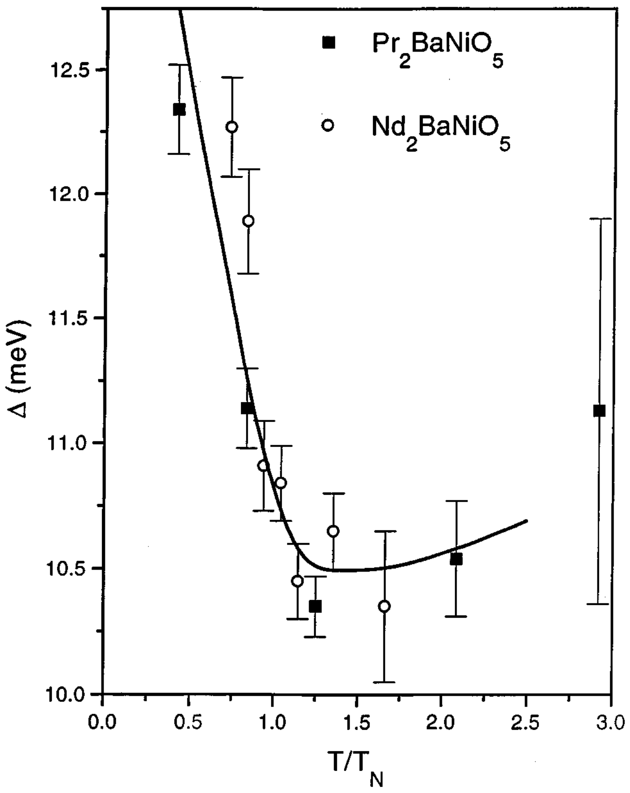

FIG. 3. Temperature dependence of the Haldane gap measured in the Nd_2BaNiO_5 powder sample (open circles, $T_N=48$ K) and Pr_2BaNiO_5 single crystal (solid squares, $T_N=24$ K). The solid line is a guide to the eye.

Y_2BaNiO_5 (Ref. 14). The data were analyzed using Eqs. (5), (6), and (8) convoluted with the resolution function. The corresponding fits are represented by solid lines in Fig. 4. For Δ we have utilized the values deduced from constant-Q scans. The other parameters appeared to be of small relevance and the values measured previously for Pr_2BaNiO_5 were used ($\kappa=0.08$ Å^{-1} and $c_0=200$ meV Å). Very good fits were obtained with the overall scaling factor and flat background being the only adjustable parameters. The dashed line in the lower panel of Fig. 4 shows the powder dynamic structure factor calculated with $P(z)=(1-z^2)$, i.e., for spin fluctuations parallel to the chain direction. This profile is clearly inconsistent with the experimental data.

C. Crystal-field excitations

As mentioned above, we have observed several CF excitations in Nd_2BaNiO_5 powder. These modes appear as symmetric Gaussian-shaped peaks in constant-Q scans (Fig. 5, insets), centered at ≈23, 18, and 4 meV (at $T=10$ K). Measurements at different momentum transfers in the range 1–5 Å^{-1} revealed no Q dependence of the peak positions or energy widths. The energy-integrated intensity of all three features shows a slow decrease at large Q, consistent with the Nd^{3+} magnetic form factor, although the amount of the data is not sufficient for a detailed form-factor analysis. The two higher-energy excitations are only weakly T dependent, their energy decreasing slightly on warming from 12 to 80 K (Fig. 5) and the integrated intensity remaining constant. In contrast, the low-energy CF mode is strongly temperature dependent and could only be observed in the ordered phase. The excitation energy increases rapidly with decreasing T, leveling off below $T\approx 20$ K. At low temperature the corresponding inelastic peak is resolution limited. However, as $T\rightarrow T_N$ the energy width increases (open circles in Fig. 5)

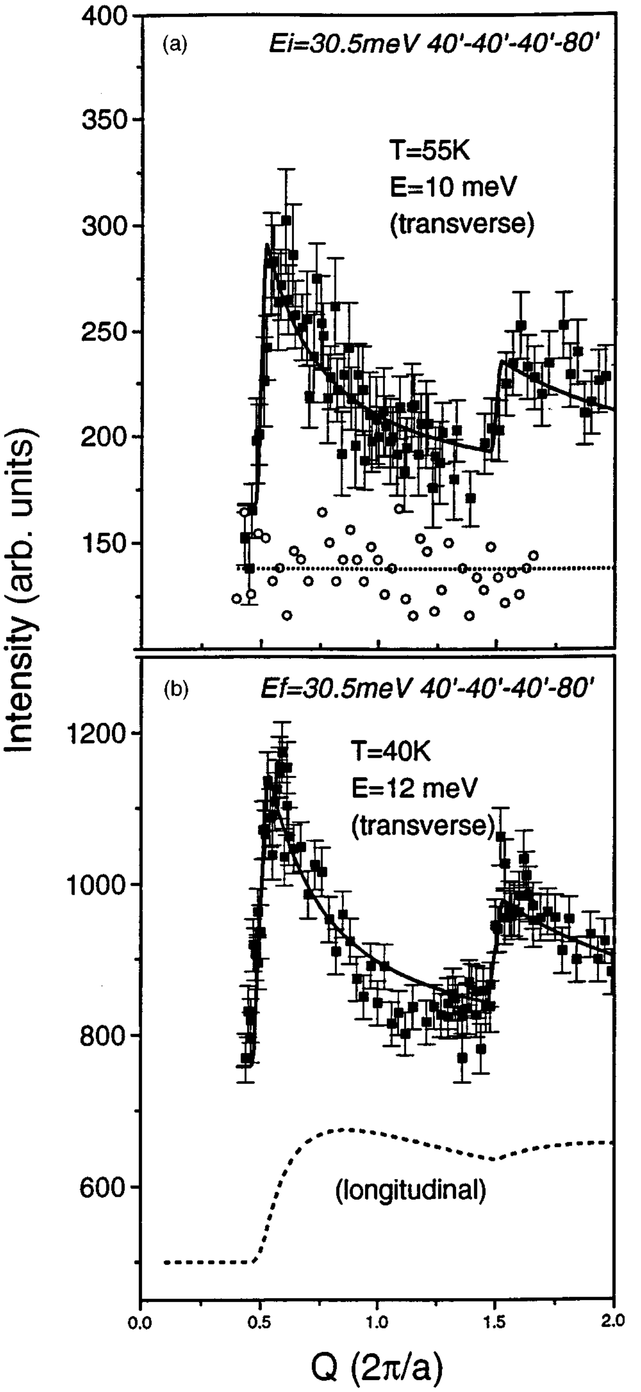

FIG. 4. Constant-E scans for the Nd_2BaNiO_5 powder collected above (a) and below (b) the Néel temperature $T_N=48$ K. The solid lines show fits with the powder cross section for transverse Haldane gap modes derived in the text. The open circles indicate the background measured below the gap ($\hbar\omega=6$ meV). The dashed line represents the powder dynamic structure factor calculated for longitudinal modes.

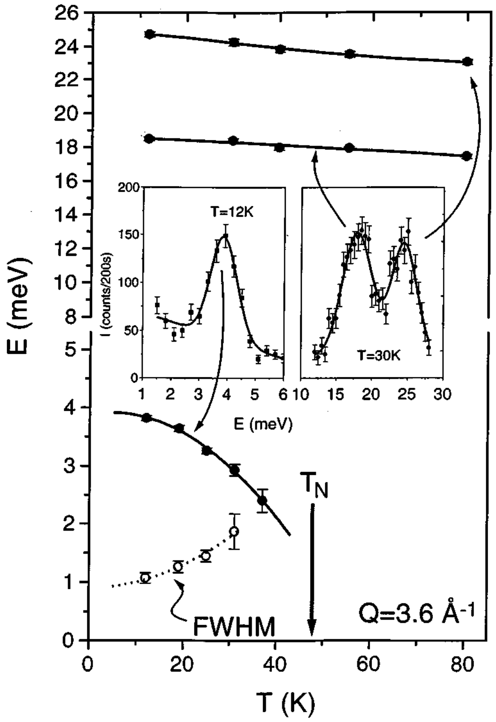

FIG. 5. Temperature dependence of the crystal-field (CF) excitations in Nd_2BaNiO_5 (solid circles) and the observed FWHM of the lowest-energy CF mode (open circles). Typical constant-Q scans (insets) are fitted with Gaussian profiles (solid lines).

and the mode becomes overdamped. The finite experimental energy resolution makes it impossible to determine whether or not the mode actually undergoes a complete softening at T_N. In the entire temperature range 10–35 K, where this low-energy excitation is observable, the energy-integrated intensity was found to be constant.

Since the main purpose of this study was the investigation of the 1D Ni-chain modes, we have not performed a crystal-field analysis of the observed CF excitations. In fact, the site symmetry ($mm2$) for Nd^{3+} in Nd_2BaNiO_5 is sufficiently low to split the tenfold degenerate $J=9/2$ multiplet into five Kramers doublets. A multitude of CF transitions can therefore be expected (see, for example, Ref. 23), and more measurements in a wider energy range are required to get the full picture.

IV. DISCUSSION

1D magnetic gap excitations are seen in Nd_2BaNiO_5 both above and below the 3D ordering temperature at ≈11 meV. Below T_N the gap steadily increases with decreasing T. In this respect the behavior is very similar to that recently observed in single-crystal inelastic neutron-scattering experiments on Pr_2BaNiO_5 (Ref. 12). For the latter compound it was shown that the gap modes preserve their ''Ni-only'' nature and purely 1D dispersion relation below T_N. However, due to the limitations in the experimental geometry and sample size, a reliable polarization analysis of the ≈11 meV mode could not be performed for Pr_2BaNiO_5. The powder structure factor $S_{\mathrm{pow}}(Q)$ measured in Nd_2BaNiO_5 resolves the ambiguity, showing that the gap mode is a transverse one. Another important point is that long-range ordering in Pr_2BaNiO_5 is driven by a soft magnetic-exciton band, the magnetic moment on Pr being quenched by the low-symmetry crystal field above T_N. This is not the case in Nd_2BaNiO_5: Nd^{3+} is a Kramers ion and always carries a nonzero magnetic moment. The difference in ordering mechanisms in the two related systems apparently does not affect the Ni-chain excitations significantly.

The existence of Haldane-like excitations in quasi-1D compounds that exhibit 3D AF order at low temperatures is not in itself a new finding. Indeed, $CsNiCl_3$, the first compound in which the Haldane gap was observed experimentally, orders antiferromagnetically below $T_N=4.8$ K (Refs. 16, 17, and 19). Haldane-like features were also observed in related integer-spin systems $RbNiCl_3$ and $CsNiBr_3$.[18,8] In these materials however the 3D dispersion in the gap modes is strongly temperature dependent and as $T \rightarrow T_N$ a complete softening occurs at the 3D AF point.[24] In the 3D phase these two branches are transformed into conventional *gapless* 3D antiferromagnons (Goldstone modes). In contrast, the gap modes in Nd_2BaNiO_5 persist in the ordered phase as purely 1D excitations. The similarity between Nd_2BaNiO_5 and the $ANiCl_3$ systems is that at the *1D antiferromagnetic zone center* the gap shows no softening at T_N.[19] As mentioned above, the Haldane-like gap excitations in L_2BaNiO_5 represent spin fluctuations on the Ni sites only.[12] Conventional AF spin waves in L_2BaNiO_5, on the other hand, must have comparable contributions from the rare earth as well as Ni spins, since 3D ordering produces a finite magnetic moment on both sites. The observed gap excitations in the ordered phase of L_2BaNiO_5 are therefore distinct from acoustic magnons.

The central question is whether the Ni-chain gap excitations in the 3D phase are a purely quantum effect related to the integrity of Ni^{2+} spins, or merely another ''optical'' branch of conventional 3D spin waves. The similarity of the behavior observed at $T>T_N$ in Nd_2BaNiO_5 to that seen in the quantum-disordered Y_2BaNiO_5 suggests the former. The same conclusion is supported by the fact that even at low temperatures the dynamic structure factor is very similar to the one calculated for a Haldane-gap antiferromagnet. To obtain a more convincing proof, spin-wave theoretical calculation of the magnetic excitation spectrum must be performed and the results directly compared to our neutron-scattering data on Nd_2BaNiO_5 and Pr_2BaNiO_5. On the experimental side, further insight may be obtained by performing field-dependent and polarized-neutron measurements and determining the multiplicity and eigenvectors of the Ni-chain modes. These approaches have been very successful in revealing the purely quantum origin of gap excitations in the 3D ordered phases of $CsNiCl_3$ and related $S=1$ compounds,[8,17–19,25] as opposed to ''classical'' modes seen in isostructural half-integer spin systems.[26–28] Our results for Nd_2BaNiO_5 clearly indicate that the gap excitation corresponds to fluctuations of spin components perpendicular to the chain direction in the crystal, but it remains unclear whether the mode is a doublet (corresponding to the two transverse Haldane modes) or not. Moreover, the third component of the Haldane triplet (seen at $\hbar\omega \approx 17$ meV in Y_2BaNiO_5), if present, could not be observed in Nd_2BaNiO_5, since it is very close in energy to one of the stronger CF peaks. These remaining ambiguities can be resolved if large single crystals become available for neutron-scattering experiments.

An interesting observation is that according to neutron diffraction[15] and resonant magnetic x-ray scattering[29] experiments the Ni^{2+} moments in $\mathrm{Nd_2BaNiO_5}$ become saturated very rapidly below $T_N = 48$ K and are practically temperature independent below $T = 40$ K. However, the observed energy of the Ni-chain modes continues to increase and their spectral weight to decrease way below this point. This effect coexists with a steady increase in the Nd^{3+} ordered moment, which grows linearly with decreasing temperature between 40 and 10 K.[15,29] It is feasible that the temperature dependence in the Ni-chain excitations is due to the influence of an effective temperature-dependent staggered field induced by the increasing rare-earth magnetic moments.

We would like to point out that, to our knowledge, there has not been so far any theoretical studies of the temperature-dependent spin dynamics in *mixed-spin* linear-chain quantum antiferromagnets like $L_2\mathrm{BaNiO_5}$, $L \neq \mathrm{Y}$. Further theoretical input and more experimental work are required to gain a complete understanding of the fine interplay between 1D and 3D phenomena.

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[1] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983).

[2] F. D. M. Haldane, Phys. Rev. Lett. **50**, 1153 (1983).

[3] O. Golinelli, T. Jolicoeur, and R. Lacaze, Phys. Rev. B **45**, 9798 (1992).

[4] O. Golinelli, T. Jolicoeur, and R. Lacaze, Phys. Rev. B **46**, 10 854 (1992).

[5] O. Golinelli, T. Jolicoeur, and R. Lacaze, J. Phys. Condens. Matter **5**, 1399 (1993).

[6] S. Ma *et al.*, Phys. Rev. Lett. **69**, 3571 (1992).

[7] L. P. Regnault, I. Zaliznyak, J. P. Renard, and C. Vettier, Phys. Rev. B **50**, 9174 (1994).

[8] K. Kakurai, Physica B **180-181**, 153 (1992).

[9] D. J. Buttrey, J. D. Sullivan, and A. L. Rheingold, J. Solid State Chem. **88**, 291 (1990).

[10] E. Garcia-Matres *et al.*, J. Solid State Chem. **103**, 322 (1993).

[11] T. Yokoo, T. Sakaguchi, K. Kakurai, and J. Akimitsu, J. Phys. Soc. Jpn. **64**, 3651 (1995).

[12] A. Zheludev, J. M. Tranquada, T. Vogt, and D. J. Buttrey, Phys. Rev. B **54**, 6437 (1996).

[13] J. Darriet and L. P. Regnault, Solid State Commun. **86**, 409 (1993).

[14] J. F. DiTusa *et al.*, Physica B **194**, 181 (1994).

[15] V. Sachan, D. J. Buttrey, J. M. Tranquada, and G. Shirane, Phys. Rev. B **49**, 9658 (1994).

[16] W. J. L. Buyers *et al.*, Phys. Rev. Lett. **56**, 371 (1986).

[17] R. M. Morra, W. J. L. Buyers, R. L. Armstrong, and K. Hirakawa, Phys. Rev. B **38**, 543 (1988).

[18] Z. Tun, W. J. L. Buyers, A. Harrison, and J. A. Rayne, Phys. Rev. B **43**, 13 331 (1991); W. J. L. Buyers, Z. Tun, A. Harrison, J. A. Rayne, and R. M. Nicklow, Physica B **180-181**, 222 (1992).

[19] K. Kakurai, M. Steiner, R. Pynn, and J. K. Kjems, J. Phys. Condens. Matter **3**, 715 (1991).

[20] S. W. Lovesey, *Theory of Neutron Scattering from Condensed Matter* (Clarendon, Oxford, 1984), Vol. 2.

[21] K. Nomura, Phys. Rev. B **40**, 2421 (1989).

[22] S. V. Meshkov, Phys. Rev. B **48**, 6167 (1993).

[23] A. Furrer, P. Allenspach, J. Mesot, and U. Staub, Physica C **168**, 609 (1990).

[24] I. Affleck, Phys. Rev. Lett. **62**, 474 (1989).

[25] M. Enderle, K. Kakurai, M. Steiner, and H. Weinfurter, J. Magn. Magn. Mater. **104-107**, 809 (1992).

[26] A. Harrison, M. F. Collins, J. Abu-Dayyeh and C. V. Stager, Phys. Rev. B **43**, 679 (1991).

[27] M. Enderle, K. Kakurai, K. N. Clausen, T. Inami, H. Tanaka, and M. Steiner, Europhys. Lett. **25**, 717 (1994).

[28] T. Inami, K. Kakurai, H. Tanaka, M. Enderle, and M. Steiner, J. Phys. Soc. Jpn. **63**, 1530 (1994).

[29] A. Zheludev, J. Hill, and D. J. Buttery, following paper, Phys. Rev. B **54**, 7216 (1996).