# Polarized-Neutron Observation of Longitudinal Haldane-Gap Excitations in $Nd_2BaNiO_5$

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Polarized and unpolarized inelastic neutron scattering is used to study Haldane-gap excitations in the mixed-spin linear-chain antiferromagnet $Nd_2BaNiO_5$. The longitudinal mode, polarized along the direction of ordered moments, is observed for the first time. The model of isolated Haldane chains in a static staggered exchange field, that is known to work very well for static properties and transverse spin excitations in $R_2BaNiO_5$ compounds, fails to explain new results for the longitudinal spin gap. [S0031-9007(99)08679-2]

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The novel quantum-disordered ground state and the famous Haldane gap in the magnetic excitation spectrum [1] have kept integer-spin one-dimensional (1D) Heisenberg antiferromagnets (AF) at the center of attention of condensed matter physicists for almost two decades. Among the more recent developments are studies of such systems in external magnetic fields (a comprehensive bibliography can be found in Refs. [2–4]). As the uniform magnetic field $H$ is increased, the Haldane triplet splits linearly with $H$, with one of the three excitations decreasing in energy and eventually reaching zero at some critical field $H_c$ [5,6]. The effect of a strong *staggered* field $H_\pi$, to which the Haldane chain is most susceptible, is expected to be no less dramatic. According to recent theoretical results [7,8] and numerical simulations [9], all three Haldane gap energies *increase* quadratically with $H_\pi$. The degeneracy of the Haldane triplet is partially lifted: for the excitation polarized along the direction of induced moments (longitudinal mode) the gap increases three times more rapidly than for the two transverse modes. The *longitudinal* mode is of particular interest, as it is a purely quantum feature, totally absent in the classical spin wave theory that predicts a pair of transverse order-parameter excitations (magnons).

The discovery of coexistence of Haldane gap excitations and magnetic long-range order in $R_2BaNiO_5$ ($R$ = magnetic rare-earth) compounds [10–14] presented a unique opportunity to investigate experimentally the effect of a strong staggered field on a Haldane spin chain. In $R_2BaNiO_5$ the initially quantum-disordered AF $S = 1$ $Ni^{2+}$ chains become subject to an effective staggered exchange field produced by the $R$ sublattice, when the latter orders magnetically at low temperatures. The magnitude of this field can be tuned by varying the temperature and thereby the ordered moment on the $R^{3+}$ sites. A wealth of neutron scattering data for a number of $R_2BaNiO_5$ compounds, particularly $Pr_2BaNiO_5$ [10] and $(Nd_xY_{1-x})_2BaNiO_5$ [11,12], have been accumulated to date. The quadratic increase in the gap energy in the magnetically ordered state has been clearly observed [10–12], and the measured temperature dependencies of sublattice magnetizations [13,15] were shown to be quantitatively consistent with predictions for isolated Haldane chains in a staggered field [8,9]. Surprisingly, the *splitting* of the Haldane triplet has never been found. In fact, no clear evidence for the very *existence* of the longitudinal mode in $R_2BaNiO_5$ has been obtained: all the measured staggered field dependencies of the Haldane gap energies are in *quantitative* agreement with calculations for *transverse* excitations in isolated chains [7–9,13]. The present paper is aimed at resolving this puzzle. We report the first direct experimental observations of the longitudinal mode in a $R_2BaNiO_5$ compound using spin-polarized inelastic neutron scattering. In the ordered phase we find that the theory of an isolated chain in a *static* staggered field, that seems to apply so well to transverse Haldane excitations in $R_2BaNiO_5$, does *not* work for the longitudinal mode.

The material of choice for our studies was $Nd_2BaNiO_5$, one of the few "2115" nickelates for which high-quality single crystals can be prepared. The orthorhombic crystal structure of $R_2BaNiO_5$ compounds is discussed in great detail elsewhere [16] and we only note here that the $S = 1$ $Ni^{2+}$ chains run along the $a$ axis of the crystal (Ni-Ni spacing $a = 3.85$ Å), with the $R^{3+}$ sites positioned in between these chains. The Néel temperature for $Nd_2BaNiO_5$ is $T_N = 48$ K. Long range magnetic ordering gives rise to magnetic Bragg reflections of the type $(\frac{2m+1}{2}, k, \frac{2n+1}{2})$, with $m$, $k$, and $n$ integer [17,18]. The $Ni^{2+}$ moments are confined in the $(a, c)$ crystallographic plane and are aligned roughly along the $c$ axis [17,19].

In previous studies the attempts to determine the polarization of the Ni-chain spin excitations in $R_2BaNiO_5$ were performed using unpolarized neutrons. In that type of experiment, for scattering vectors almost along the chain direction (crystallographic $a$ axis) one sees the fluctuation of spin components only along the $b$ and $c$ axes of the

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crystal, thanks to the intrinsic polarization dependence of the neutron scattering cross section. When the scattering vector is almost perpendicular to the chain axis and almost parallel to $c$, for example, only the $a$- and $b$-spin components contribute to scattering. In principle, comparing the intensities measured at several wave vectors can yield a complete analysis of the mode polarization. In practice, however, such measurements on $Nd_2BaNiO_5$ were inconclusive [10,13]. The main difficulty is that a number of intrinsic effects, particularly neutron absorption in the sample and focusing, are very difficult to account for with sufficient accuracy when comparing measurements for a large irregularly shaped sample and substantially different scattering vectors.

Even if a polarization analysis could not be properly performed in the above-mentioned experiment, a splitting of the triplet, if present at all, should have been seen. Our first guess was that the mode splitting was obscured by the relatively low energy resolution (2.6 meV FWHM at 15 meV energy transfer) of previous experiments [13]. As a first step in clarifying the behavior of the longitudinal mode we therefore performed additional unpolarized measurements at the High Flux Isotope Reactor at Oak Ridge using a high-resolution setup that employed a Be(002) monochromator, a PG(002) analyzer, and $60'$-$40'$-$40'$-$240'$ collimations to yield a 1.6 meV FWHM resolution at 15 meV energy transfer (fixed final energy 14.7 meV). In these studies we used the same large sample as in Ref. [13]. A typical inelastic constant-$Q$ scan through the Haldane gap excitations at the 1D AF zone center $\boldsymbol{Q} = (1.5, 0, 0)$ is shown in Fig. 1. The background was measured and subtracted as in Ref. [13]. From the prediction for an isolated chain [8], we expect the longitudinal mode to appear at $\approx$19 meV energy transfer at this temperature ($T = 38$ K), whereas no such feature is observed. In fact, none of our inelastic scans performed in the temperature range 30–55 K revealed any splitting of the triplet, in agreement with low-resolution studies [13].

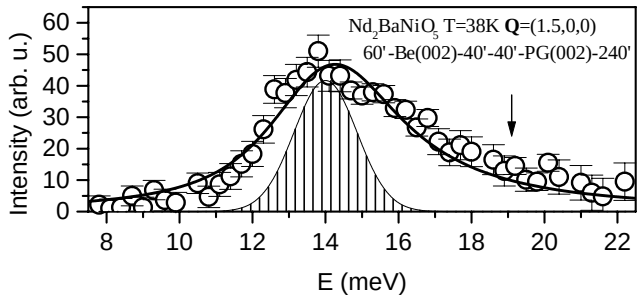


FIG. 1. A typical inelastic scan measured in $Nd_2BaNiO_5$ with unpolarized neutrons. The background has been subtracted from the data. The shaded Gaussian represents the experimental energy resolution. The solid line is a simulation based on Eq. (2) and parameters determined in a polarized-neutron experiment. The arrow shows the position of a second inelastic peak predicted by the static-staggered-field model.

To finally resolve the mystery of the longitudinal mode we made use of a totally different technique to measure the polarization of the magnetic gap excitations in $Nd_2BaNiO_5$. Employing a polarized neutron three-axis setup [20] we performed all the measurements at a *single* position in reciprocal space to avoid any complications of absorption or focusing. Magnon polarization was determined by comparing the inelastic intensity measured with different combinations of incident and final neutron polarizations. The experiment was carried out at the IN-20 polarized neutron spectrometer at the Institut Laue-Langevin in Grenoble. The sample that was previously used in unpolarized experiments was mounted on the spectrometer with the $c$ axis vertical. The induced moment on the Ni chains is in the $(a, c)$ plane and forms an angle of $\phi = 30°$ with the $c$ axis (the vertical direction). A combination of Heussler-alloy monochromator and analyzer and two Mezei-type flippers allowed us to polarize and analyze the incident and outgoing beams parallel or antiparallel to the vertical axis at will. The measured flipping ratio for each flipper was approximately 21. In this geometry, in the non-spin-flip (NSF) channel one sees *only* the fluctuation of spin components along the vertical $c$ axis. The largest fraction ($\cos^2\phi = 0.75$) of this intensity is due to spin fluctuations along the direction of staggered field, while the remaining 25% correspond to transverse spin fluctuations. In contrast, in a spin-flip (SF) configuration only the $a$- and $b$-axis spin components are seen. The experiment was done using 14.7 meV fixed-final energy neutrons with a PG (pyrolytic graphite) filter positioned after the sample. All constant-$Q$ scans were collected at $\boldsymbol{Q} = (1.5, 0.5, 0)$, which corresponds to a wave vector transfer $\boldsymbol{Q}_\parallel = 3\pi/a$ along the Ni-chain axis. For the particular scattering geometry in the SF channel 21% and 79% of the intensity is due to $a$- and $b$-axis spin components, respectively. The slight inclination of the ordered moment away from $c$ and towards the $a$ axis leads to a negligible 5% contribution of longitudinal spin fluctuations to the measured SF intensity. The background was measured at $\boldsymbol{Q} = (1.4, 0.5, 0)$ [13]. As the background predominantly comes from crystal-field excitations associated with $Nd^{3+}$ [11] and thus has a large magnetic (polarization-dependent) contribution, it was separately measured in the NSF and SF configurations.

The main difficulty in a polarized neutron experiment is the long counting times that result from the typically low flux of spin-polarized neutrons. To use the available beam time with greatest efficiency, we concentrated on the temperature range 35–55 K where all the significant changes are expected to take place. Indeed, at the high-temperature end, $T > T_N = 48$ K, the system is in the paramagnetic phase. At $T = 35$ K, on the other hand, the $Ni^{2+}$ moments have already achieved as much as 75% of their saturation value ($\approx 1.1\mu_B$) [13,15]. The bulk of our polarized-neutron inelastic data (background subtracted) is shown in Fig. 2. At $T = 55$ K $> T_N$ in both SF and NSF channels, at the 1D AF zone-center $\boldsymbol{Q} = (1.5, 0.5, 0)$,

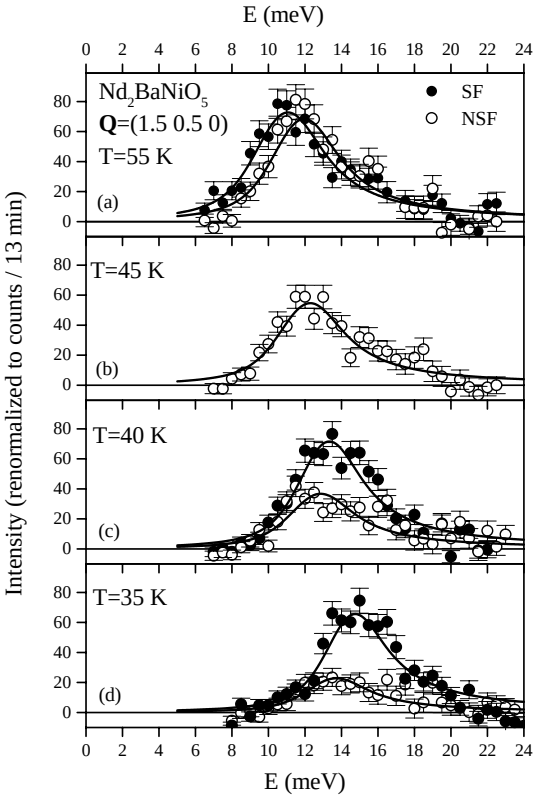


FIG. 2. Temperature evolution of constant-$Q$ scans measured in $Nd_2BaNiO_5$ using unpolarized neutrons. Open and solid circles correspond to spin-flip and non-spin-flip scattering. The solid lines are fits to the data with based on Eq. (2).

we clearly see two well-defined inelastic peaks of almost equal intensity, centered at 11 and 12 meV, respectively [Fig. 2(a)]. As the temperature is decreased through $T_N$, both peaks move to higher energies [Figs. 2(b)–2(d)]. The peak observed in the NSF configuration (longitudinal mode), if anything, moves to higher energies *more slowly* than the SF peak (transverse modes). The intensity of the NSF peak decreases rapidly in the magnetically ordered phase. At $T = 35$ K the longitudinal mode is practically gone: most of the observed NSF intensity (75%) is due to transverse excitations and the slight canting of the staggered field, as explained above. The intensity in the transverse modes appears to be practically temperature independent.

To make the above discussion more quantitative we analyzed our inelastic scans with the following one-dimensional cross section that is routinely employed to model the dynamic structure factor of Haldane excitations:

$$(\hbar\omega_q)^2 = c_s^2 q_\parallel^2 + \Delta^2, \tag{1}$$

$$S(q_\parallel,\omega) = \frac{A c_s}{\Gamma\Delta}\left(1 + \left[\frac{c_s(q_\parallel - \pi/a)}{\Delta}\right]^2 + \left[\frac{\omega - \omega_q}{\Gamma}\right]^2\right)^{-1}. \tag{2}$$

Here $c_s = 210$ meV · Å is the spin wave velocity measured with great accuracy using unpolarized neutrons [21], $\Delta$ is the Haldane gap energy, $a$ is the chain lattice constant, and $\Gamma$ is the intrinsic energy width of the excitation. The prefactor $A$ is proportional to the energy-integrated intensity. The dynamic structure factor (2) was numerically convoluted with the spectrometer resolution function. For $\Gamma$ we used the same values as in Ref. [13], derived from previous $\Gamma(T)$ measurements on $Y_2BaNiO_5$ [22]. The parameters $\Delta$ and $A$ were then refined to best fit each scan. The resulting curves fall on the data points rather well and are shown as solid lines in Fig. 2. In Fig. 3 (large open and solid circles) we plot the temperature dependence of the gap energies and integrated intensities that characterize the NSF and SF peaks.

It is important to make sure that our new findings are consistent with unpolarized neutron scattering results. The temperature dependence of the gap energy measured previously at $\boldsymbol{Q} = (1.5, 0, 0)$ with unpolarized neutrons (Ref. [13]) is plotted in small triangular symbols in Fig. 3(a). For the intensity of the excitations we have to

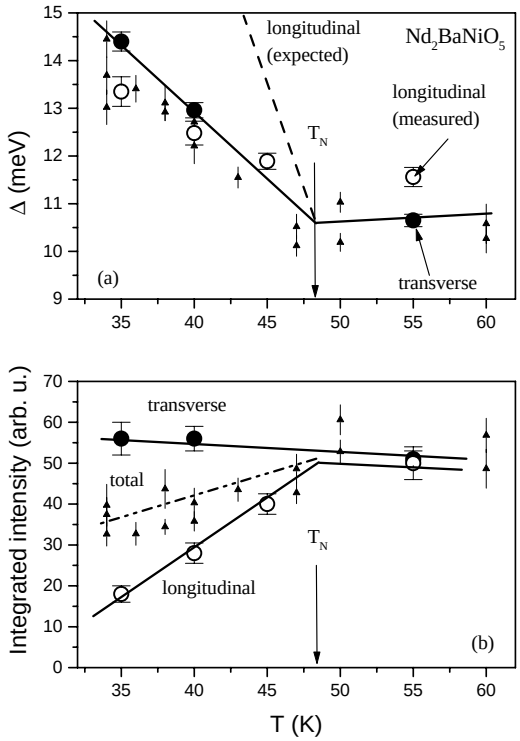


FIG. 3. (a) Measured temperature dependence of the energy gap in the longitudinal (open circles) and transverse (solid circles) Ni-chain excitations in $Nd_2BaNiO_5$. The solid lines are guides for the eye. The dashed line is the theoretical prediction for the longitudinal mode in the static staggered field model [8,13]. Small triangles show the data previously obtained with unpolarized neutrons [8,11,13]. (b) Measured temperature dependence of the energy-integrated intensity of the longitudinal (open circles) and transverse (solid circles) Ni-chain modes in $Nd_2BaNiO_5$. The lines are guides for the eye. Small triangles are as in (a).

compare the *averaged* SF and NSF intensities measured in our experiments [dash-dotted line in Fig. 3(b)] to the intensity measured without polarization analysis [Fig. 3(b), small solid triangles]. Finally, using the parameter values obtained through fitting Eq. (2) to the polarized neutron scans we can simulate unpolarized scans (an example is shown solid line in Fig. 1). We see that within the statistical scattering in the data points our new results agree very well with all existing unpolarized data.

The most important result of the present study is the unambiguous evidence for the existence of the longitudinal mode and its persistence, at least over an appreciable temperature range, in the magnetically ordered state. Now there remains little doubt that the Ni-chain excitations in $Nd_2BaNiO_5$ are indeed a Haldane triplet. In fact, at $T > T_N$, the intensity of the longitudinal and transverse modes are practically equal, as in the case of an isolated Haldane spin chain. In the paramagnetic phase there seems to be little difference between $Nd_2BaNiO_5$ and the reference Haldane system $Y_2BaNiO_5$, as previously suggested by the measured temperature dependence in mixed $(Nd_xY_{1-x})_2BaNiO_5$ compounds [12]. For example, just as in $Y_2BaNiO_5$ [23], our values for the $c$- and $b$-axis gaps in $Nd_2BaNiO_5$ are different by roughly 1 meV. This initial splitting of the triplet is believed to be a result of the weak single-ion anisotropy on the $Ni^{2+}$ sites.

Another important finding is that the intensity of transverse excitations in $Nd_2BaNiO_5$ is not affected by magnetic ordering. The previously observed decrease of intensity of the Haldane excitations in $R_2BaNiO_5$ [13] below $T_N$ is thus entirely due to a suppression of the longitudinal mode. An overall decrease of inelastic intensity is of course to be expected: in the ordered state a substantial amount of the spectral weight associated with $Ni^{2+}$ moments is transferred from dynamic to static spin correlations, i.e., into the magnetic Bragg reflections.

A totally new and yet unexplained result of our polarized neutron studies is the relatively slow increase of the longitudinal gap seen upon cooling through the ordering temperature. Indeed, for an isolated chain in a *static* staggered field the longitudinal gap $\Delta_\parallel$ is expected to increase with decreasing $T$ three times as fast as $\Delta_\perp$ [7–9], as indicated by the dashed line in Fig. 3(a). This discrepancy with the static field model means that one cannot consider the Ni and Nd magnetic degrees of freedom to be totally independent, at least for longitudinal fluctuations. The longitudinal mode apparently involves both the Ni and Nd moments, and may also be coupled with higher-energy crystal-field levels of the rare earths. A further investigation of this problem is required. In particular, a more detailed RPA analysis of the interactions between the Ni- and rare-earth subsystems would be very useful.

In summary, we have for the first time observed the longitudinal Ni-chain excitation in a $R_2BaNiO_5$ system. We have shown that the "Haldane chain in a staggered field" model may be a good starting point, but fails to account for all the details of the spin excitation spectrum in these remarkable mixed-spin quantum magnets.

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