Direct evidence for nuclear spin waves in Nd$_2$CuO$_4$ by high-resolution neutron-spin-echo spectroscopy

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Abstract

The possibility of coupling through the hyperfine interaction of nuclear spins with the electronic spin system has given rise to hope for potential novel applications in spintronics and quantum computations. We investigated the dispersion of nuclear spin waves in such a coupled system, Nd$_2$CuO$_4$, by using neutron-spin-echo spectroscopy at millikelvin temperatures. Our results show the existence of dispersion of nuclear spin waves in Nd$_2$CuO$_4$ at $T = 40$ mK. A fit of the dispersion data with the spin wave dispersion formula gave the Suhl–Nakamura interaction range to be of the order of 10 Å, which is much smaller than that expected theoretically.

(Some figures may appear in colour only in the online journal)

1. Introduction

The coupling of nuclear spins with the electronic spin system through the hyperfine interaction has attracted a great deal of interest due to its possible application in spintronics and quantum computations [1, 2]. The coupling of nuclear spins through the Suhl–Nakamura indirect interaction gives rise to nuclear spin excitations [3]. Each nuclear spin sees the electronic spin on its own ion through the effective hyperfine coupling $A \mathbf{I} \cdot \mathbf{S}$, where $A$ is the hyperfine parameter, $\mathbf{I}$ is the nuclear spin and $\mathbf{S}$ is the electronic spin. The electronic spins of all the ions are coupled by exchange interactions. An interaction of the nuclei therefore arises via the low-lying excited states (spin waves) of the electronic systems as intermediate states. That is to say, a nuclear spin excites a spin wave through the hyperfine coupling, and another nuclear spin causes it to be reabsorbed through its hyperfine coupling. This process gives rise to the so-called Suhl–Nakamura indirect interaction [4, 5]. Although the polarized nuclear spin system is far from being in a perfectly ordered state, it does possess long-range order of the average oriented nuclear spin because of the long range of the Suhl–Nakamura interactions; therefore, as pointed out and justified by de Gennes et al [6], there exists a spin wave like spectrum of excitations of the nuclear spin system. Word et al [7] calculated the relevant neutron scattering cross sections from electronic and nuclear spin systems coupled by Suhl–Nakamura interaction. There exists indirect evidence of nuclear spin waves, which cause lowering of the antiferromagnetic resonance (AFR) and nuclear magnetic resonance (NMR) frequencies at low temperatures [8]. Nuclear spin waves are expected to have energies about three orders of magnitude lower than those of electronic spin waves and should be in the $\mu$eV range. The nuclear spin wave spectrum should have dispersion at lower momentum transfer $q$-range inversely proportional to the range $b$ of the Suhl–Nakamura indirect interaction. Kurkin and Turov [9] have considered the properties of nuclear spin waves and have compared them with those of electronic spin waves. Owing to the large value of the Suhl–Nakamura interaction range there can be still a strong correlation in the motion of nuclear spins, even when they are in a disordered paramagnetic state. In the electronic case such strong correlations are seen only in low-dimensional or frustrated spin systems. There are differences in the dispersion laws for nuclear spin waves and magnons due to the difference in the coordinate dependence of the Suhl–Nakamura and...
exchange interactions. Whereas the Fourier spectrum of the short-range exchange interaction contains all the wavevectors \( k \) from the Brillouin zone, in the Fourier spectrum of the long-range Suhl–Nakamura interaction the components with \( k \) much greater than \( r_0^{-1} \) (\( r_0 \) is the Suhl–Nakamura interaction range) are sparsely represented. There is another very important difference between the properties of nuclear and electronic spin waves. The spin wave dispersions in ferro- and antiferromagnets are very different in electronic spin systems, whereas in the case of nuclear spin waves they are very similar. All effects associated with nuclear spin waves are very similar but much stronger in antiferromagnets due to the drastic difference in the Suhl–Nakamura interaction range.

There have been some NMR investigations [8, 10–13], which probe the nuclear spin waves indirectly and are essentially limited to the momentum transfer \( q = 0 \). These studies have been performed on Mn-based compounds, MnCO\(_3\), CsMnF\(_3\), CsMnCl\(_3\) and RbMnF\(_3\). The \(^{55}\)Mn nucleus (spin \( I = 5/2 \)) has a large hyperfine constant \( A = 600 \text{ MHz} \) and 100% abundance of the magnetic isotope. However, neutron scattering is the only microscopic probe that is in principle capable of measuring nuclear magnetic excitations in the \((Q, \omega)\) space. Also, in order to probe nuclear spin waves it is necessary to have very good energy as well as momentum \( q \) resolution at the same time. Neutron-spin-echo spectrometers are the only instruments that satisfy such stringent requirements. Another important factor is suitably large spin dependent scattering cross sections. Nd-compounds satisfy such requirements. Out of seven naturally occurring isotopes, \(^{143}\)Nd and \(^{145}\)Nd have spin \( I = 7/2 \) with natural abundances 12.18% and 8.29%, respectively, and the spin dependent scattering cross sections are large. We have therefore investigated nuclear spin excitations in the well-known parent compound Nd\(_2\)CuO\(_4\) of the electron-doped superconducting compounds Nd\(_2\)−xCe\(_x\)CuO\(_4\).

A large well-characterized single crystal of Nd\(_2\)CuO\(_4\) was available to us. Hyperfine induced nuclear spin ordering in Nd\(_2\)CuO\(_4\) below about 200 mK has been reported by Chattopadhyay and Siemensmeyer [14] from their neutron diffraction measurements. Nd\(_2\)CuO\(_4\) is therefore a very good candidate for observing spin dynamics of a nuclear system that is ordered at attainable low temperature. To the best of our knowledge this is the only nuclear spin system that is known to order via hyperfine interaction below about 200 mK. Chatterji and Frick [15] have reported the observation of nuclear spin excitations in Nd\(_2\)CuO\(_4\) from their inelastic neutron scattering investigation with a back-scattering spectrometer. However, the dispersion of the nuclear spin waves that is expected to occur in a very small \( q \) range could not be measured because the back-scattering spectrometer did not have the required \( q \) resolution.

So far there have been no direct experimental investigations on nuclear spin waves and their properties. The theoretical predictions described before need to be checked by suitable experiments. A propagating spin wave is characterized by its finite dispersion and to establish its existence it is necessary to measure the dispersion of its energy as a function of the momentum transfer. The only available direct experimental technique for measuring the dispersion of nuclear spin waves is high-resolution inelastic neutron scattering. In order to establish experimentally the existence of nuclear spin waves and their various properties derived theoretically and described above, we have undertaken inelastic neutron scattering experiments by using a neutron-spin-echo spectrometer that has the required energy and momentum resolution. We have successfully measured the dispersion of nuclear spin waves in Nd\(_2\)CuO\(_4\) in the hyperfine induced ordered nuclear magnetic phase at millikelvin temperatures and we describe and discuss the results of this investigation in this paper.

2. Experimental procedure

The experiment was carried out at the J-NSE spectrometer [16, 17] at the FRM II, Garching. The sample was mounted in a Kelvinox cryostat with a \(^3\)He–\(^4\)He dilution insert and was cooled down to 40 mK. The sample temperature was maintained at 40 mK throughout the experiment. The J-NSE was equipped with a rotating sample stage and the second arm could be turned in the horizontal plane with the origin of rotation at the sample position. However, it was not possible to tilt the sample due to the absence of tilting goniometers in the spectrometer. Therefore the crystal was pre-oriented with its \([1, 1, 0]\) crystallographic axis vertical such that the scattering plane was \((hhl)\), enabling one to access the nuclear magnetic Bragg peak at \(\{\frac{1}{2}, \frac{1}{2}, 0\}\) and measure the dispersion along \([001]\). Attempts to measure the nuclear spin wave dispersion along \([110]\) did not succeed due to the unfavorable instrumental \(q\) resolution of the spectrometer along this direction. Figure 1 shows schematically the reciprocal scattering \(hhl\) plane and scan directions. The beam was collimated by the distance between the neutron guide exit (60 × 60 mm\(^2\)) and the sample (10 × 10 mm\(^2\)) and had a divergence of about 0.5°. A 10% velocity selector provided a neutron beam with a wavelength of 5 ± 0.25 Å. A two-dimensional \(^3\)He-multidetector at a distance of 4.33 m from the sample accepted an angular range of ±2° from the central point. The normalized intermediate scattering function \(S(q, \tau)/S(q, 0)\) was measured in the standard configuration (including the \(\pi\)-flipper) [17]. Here \(\tau\) is the Fourier time.
Figure 2. $S(q, \tau)/S(q, 0)$ as a function of the Fourier time $\tau$ at $Q = (1/2, 1/2, 0)$, $(1/2, 1/2, 0.1)$, $(1/2, 1/2, 0.2)$ and $(1/2, 1/2, 0.4)$ with the least-squares fit results with a sine function.

A reference measurement with an elastic scatterer (a stochastic TiZr alloy) was carried out for $|q| = |1/2 1/2 0|$ and the same set of Fourier times as was measured from the sample. This measures the instrumental resolution. Compared to measurements of $S(q, \omega)$, where resolution corrections involve a Fourier transform (unfolding of the spectra with the instrumental resolution function), this correction is a simple division in neutron-spin-echo spectroscopy due to the intrinsic Fourier transform $S(q, \tau)/S(q, 0)$ of the NSE technique. The $q$-resolution in the direction of $\vec{q}$ is determined by the divergence of the beam and the wavelength spread $\Delta \lambda / \lambda = 0.1$, while perpendicular to it only the beam divergence gives the major contribution. The elongated instrumental resolution favors measurements in the $[1, 1, X]$-direction. The oscillations measured in this experiment always started at a minimum, which indicates that the resulting scattered intensity is the sum of an elastic non-spin-flip and the oscillating spin-flip processes. The ratio of the two determines the $y$-axis offset of the oscillation.

The spin-flip scattering flips all spin directions (like the $\pi$-flipper). The similar contributions of residual elastic coherent scattering and the magnetic scattering result in a completely depolarized elastic signal (no difference in scattered intensity whether the instrumental $\pi$-flipper is on or off). The inelastic measurements show a minimum at about $\tau = 0$. When the magnetic signal goes to the first minimum, only the residual elastic coherent contribution remains and gives rise to a maximum of the oscillation.

3. Data treatment and results

Only a limited number of scans such as those shown in figure 2 could be measured due to the long measuring times of 2–3 days per scan. The regions at the borders of the detector and the Bragg peak, if still visible on the detector, were excluded from the evaluation process of the signal. The oscillating signal could not be normalized as usual to the up- and down-intensities (all flippers off, only the...
\(\pi\)-flipper on respectively), since the mixture of spin-flip and non-spin-flip scattering erases the differences between up- and down-configurations, but not the Fourier time dependence of the signal. The non-spin-flip contribution is an elastic (i.e. constant) background arising either from residual elastic scattering of the sample or/and the cryostat Al-windows. The intermediate scattering function was fitted with the function \(S(q, \omega)/S(q, 0) = A \sin(\omega t) + B\); since NSE measures the cosine Fourier transform of the scattering function \(S(q, \omega)\), the oscillating frequency is directly related to the energy of the excitation \(E = h\omega\). Here \(A\) is the amplitude and \(B\) is the background. The nominal position of the nuclear magnetic Bragg peak, \(Q = (1/2, 1/2, q)\), was set to the maximum intensity of the peak after the \(\theta = 2\theta\)-scan. Measuring \(S(q, \tau)/S(q, 0)\) at this position and evaluating the detector in the vicinity of the peak lead to a very noisy and weak oscillation; hence large errors are encountered for this setup. On moving away from the peak by rotating the sample, an oscillation could be clearly identified with a frequency of about \(f = 2.6 \text{ ns}^{-1} (h\omega = 1.7 \text{ \mu eV})\) where \(\omega = 2\pi f\). This oscillation frequency was rather independent of the details of the evaluation process and the exact size of the patch on the detector that was evaluated. By turning the sample, one takes advantage of the higher resolution in the direction perpendicular to \(Q\), but still averages in the other direction over some \(|q|\) due to the 10\% wavelength spread, which limits the possibility to go small \(|q|\) in the dispersion relation.

Figure 2 shows \(S(q, \tau)/S(q, 0)\) as a function of the Fourier time \(\tau\) at \(Q = (1/2, 1/2, 0), (1/2, 1/2, 0.1), (1/2, 1/2, 0.2)\) and \((1/2, 1/2, 0.4)\) with the least-squares fit results with a sine function, and figure 3 shows the resulting dispersion. We fitted the dispersion with the expected dispersion for a nuclear spin system \([9]\) coupled by the Suhl–Nakamura interaction

\[
E(q) = E_s - E_p[1 + (q\tau_0)^2]^{-1}
\]

where \(E(q)\) is the energy of the nuclear spin excitation at the momentum transfer vector \(q\), \(E_s\) is the saturation value of the energy, \(E_p\) is the amount of dispersion corresponding to the frequency pulling \(\omega_0\) in the NMR experiment and \(\tau_0\) is the Suhl–Nakamura interaction range. The least-squares fit gives \(E_s = 1.76 \pm 0.5 \text{ \mu eV}, E_p = 0.12 \pm 0.05 \text{ \mu eV}\) and \(\tau_0 = 5 \pm 3\) direct lattice units. Due to the lack of sufficient data points and also due to the uncertainty of the experimental \(q\) values these fitted parameters have relatively large standard deviations.

4. Discussion

We note that the present results give the first inelastic neutron scattering evidence for the existence of nuclear spin waves. However, the results are certainly very preliminary and we still lack sufficient data points to determine the dispersion accurately. The total dispersion determined is indeed very small and is not more than 2–3 times the errors of the data points. Due to the geometrical restrictions imposed by the plate-shaped sample and due to the shape of the instrumental resolution function we mainly restricted the measurement of dispersion to only along the [001] direction. The dispersion along this direction is expected to be rather small because of the two-dimensional character of the magnetic system. The dispersion in the \(a\)-\(b\) plane, on the other hand, is expected to be much stronger. The orientation of the crystal allowed us to measure the dispersion along the [110] direction but it could not be investigated due to the worse instrumental resolution along this direction. The plate-shaped geometry of the crystal with the [001] direction perpendicular to the plate prevented us from orienting the crystal with the [001] axis vertical and measuring the dispersion along [100]. Also, due to the limited neutron beam time we could not measure the temperature dependence of the nuclear spin waves. Despite these shortcomings of the present investigation we note that the measured dispersion gives the value of the corresponding frequency pulling in the NMR experiment at \(q = 0\) to be 0.12 \text{ \mu eV}. This important result can be checked by an independent experiment using a different method, namely by the NMR technique. Unfortunately no such NMR experiment has been undertaken so far. The Suhl–Nakamura interaction range \(\tau_0\) determined by fitting the dispersion with equation (1) turns out to be 5\(c/2\pi \approx 10\) \text{ \AA}. This value is surprisingly much less than that (100 \text{ \AA} or so) expected theoretically \([6, 9]\) for the Suhl–Nakamura interaction range. This also means that the nuclear spin wave dispersion is not limited to a very small \(q\)-range close to \(q = 0\) as expected theoretically. However, the fitted value for the Suhl–Nakamura interaction range has a very large standard deviation. It is to be noted that the non-availability of tilting goniometers in the spectrometer has caused some uncertainty in the determination of the \(q\) values as well. Therefore, although we have demonstrated the existence of the dispersion of nuclear spin waves in Nd\(_2\)CuO\(_4\), the determination of the Suhl–Nakamura interaction range is not yet accurate enough. A higher \(q\)-resolution would be desirable for investigating in more detail the dispersion relation close to the nuclear magnetic Bragg peak and for the precise determination of

\[\text{Figure 3. Dispersion of the nuclear spin waves in Nd}_2\text{CuO}_4\text{ along [001]. The red curve shows the least-squares fit of the dispersion data with the nuclear spin wave model explained in the text.}\]
the Suhl–Nakamura interaction range. This could be achieved by a time-of-flight neutron-spin-echo spectrometer (e.g. the SNS-NSE [18] at SNS, Oak Ridge), where one is not limited to the 10% velocity selector for q-resolution but can adapt the resolution a posteriori by choosing the desired number of time frames for evaluation. Unfortunately, however, this otherwise very suitable neutron-spin-echo spectrometer SNS-NSE is not yet equipped with a $^3$He–$^4$He dilution cryostat, which is absolutely necessary for the success of the desired investigation. Finally, we hope that our results will prompt others to investigate this completely unexplored area of neutron science.

5. Conclusion

In conclusion, we have shown from our inelastic neutron scattering investigation with a neutron-spin-echo spectrometer the existence of dispersion of nuclear spin waves in Nd$_2$CuO$_4$ at $T = 40$ mK.

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