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Neutron Brillouin Scattering Experiments with Pulsed Neutrons on High Resolution Chopper Spectrometer HRC

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Abstract.

We improved the High Resolution Chopper Spectrometer (HRC), which is installed at MLF in J-PARC, in order to perform neutron Brillouin scattering (NBS) experiments, and successfully demonstrated a feasibility of this method. Gapless spin-wave excitations were observed in $La_{0.8}Sr_{0.2}MnO_3$, which were in good agreement with previous results using single crystals, on the other hand, a large energy gap in the ferromagnetic spin waves was found in $SrRuO_3$. Spin-wave peaks in a permanent magnet $Nd_2Fe_{14}B$ were on the dispersion curve determined previously using a single crystal. Excitations, so-called fast sound, in liquid D_2O were in a good agreement with previous inelastic neutron scattering experiments and the dispersion curve were extended to the lower Q region.

1. Introduction

Neutron Brillouin scattering (NBS) is an advantageous method for observing coherent excitations in non single-crystal materials. As a result of the powder average of the dynamical structure factor for ferromagnetic spin waves from polycrystalline samples, for instance, the scattering intensities detected in the forward direction near (000) rapidly decrease with the increase in the scattering vector Q. Owing to the kinematic constraints of neutron spectroscopy, incident neutron energy E_i in the sub-eV region is necessary for measuring scattering in the meV transferred energy E range or with a high energy resolution of $\Delta E/E_i$, further, the scattered neutrons need to be detected at very low scattering angles ϕ. Although the principles of NBS are quite old, recent efforts for realizing experimental conditions of the NBS method have been made

at spectrometers, Pharos at the pulsed spallation neutron source [1] and BRISP at the steady state neutron source [2]. We also made similar approaches on the High Resolution Chopper Spectrometer (HRC) installed at the pulsed spallation neutron source of the Material and Life Science Facility (MLF) in J-PARC (Japan Proton Accelerator Research Complex) [3]. We here demonstrate the feasibility of NBS on the HRC by some experiments.

2. NBS experiment on HRC

On the HRC [4], the monochromatic neutron beam with the energy of E_i is incident upon the sample, and the energy transfer E is determined by the time-of-flight (TOF) of the detected neutron. An array of position sensitive detectors (PSD) was installed on the HRC to detect scattered neutrons with a wide energy-momentum space. For NBS measurements [3], scattered neutrons were collected by the two-dimensional bank of the PSD array at low scattering angles $\phi = 0.5 - 2.8^\circ$. In the present experiments, the lowest angle was 0.69° due to the spread of the horizontal beam width of the direct beam. The sample was packed into a flat aluminum can. A monochromatic neutron beam was incident normal to the flat face of the sample can. The beam cross section was defined to be 3 cm $\times$ 3 cm with a cadmium plate. The accelerator was operated with approximately 200 kW of the proton beam power. The data were accumulated typically for 2 days for each condition. The intensities of the observed spectra indicated below were normalized by the accumulation time.

The reduction of background noise was essential for implementing the NBS method. A T0 chopper well cut the background noise originating from the high-energy neutron burst [4]. In order to eliminate unwanted scattering other than that from the sample, a Soller type collimator with the collimation of 0.3°, which matches the incident beam divergence, was mounted just upstream the sample for this experiment. Careful consideration of the background contamination is the most important part of the experiments, thus, we performed empty can scans.

A detector element located at ϕ scans points in the (Q, E) space is described by

$$\frac{\hbar^2 Q^2}{2m} = 2E_i - E - 2\left(E_i(E_i - E)\right)^{1/2}\cos\phi, \tag{1}$$

where $\hbar$ and m are the Planck's constant divided by 2π and the neutron mass, respectively. Low-angle detectors are essential to access the present (Q, E) space. In fact, the region above the dashed line in Fig. 1, which is $E = (\hbar^2 Q^2/2m)/\sin\phi$, or the envelope of scan loci for $\phi = 5^\circ$ in eq. (1) with respect to E_i, can never be accessed by, for instance, a conventional spectrometer with the lowest scattering angle of $\phi = 5^\circ$.

3. Nearly cubic perovskites $La_{0.8}Sr_{0.2}MnO_3$ and $SrRuO_3$

Spin waves were observed from two polycrystalline ferromagnets $La_{0.8}Sr_{0.2}MnO_3$ (Curie temperature, $T_C = 316$ K, and the saturation moment, $M_s = 3.6\ \mu_B$) [5, 6] and $SrRuO_3$ ($T_C = 165$ K and $M_s = 1.6\ \mu_B$) [7]. The sample volumes were approximately 8 and 13 cm^3, respectively. Both perovskites essentially have a cubic lattice structure with a small distortion. The former is a typical example showing a colossal magnetoresistance. The latter has recently shown an enhanced anomalous Hall effect due to the spin-orbit coupling of the Ru 4d orbital [7]. Although $SrRuO_3$ has not yet been synthesized in large single-crystal form, polycrystalline samples are readily available. The measurements were performed at temperatures $T = 6$ and 254 K for $La_{0.8}Sr_{0.2}MnO_3$ and at $T = 7$ K for $SrRuO_3$, with $E_i = 102$ meV and $\Delta E = 2.0$ meV. The background-subtracted spectra showed spin-wave peaks and an elastic peak, as shown in Figs. 1 (a) and (b). The intensities of spin waves were well explained by the temperature factor as well as the magnetic form factor. Each peak was fitted to a Gaussian function with a width determined from the resolution including the dispersion slope and the peak positions (E_p) of the spin waves were determined [3], as shown in Fig. 1 (c).

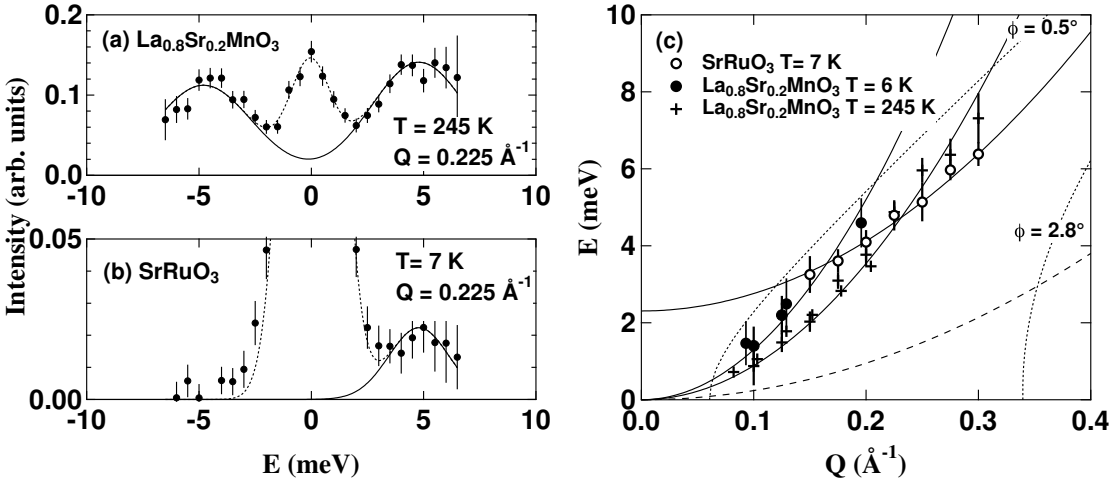

Figure 1. Typical excitation spectra for $La_{0.8}Sr_{0.2}MnO_3$ and $SrRuO_3$ (a,b). The dashed lines are fitted curves and the solid lines are the spin-wave components. Spin-wave dispersion curves (c). The solid lines are fitted dispersion curves. The dotted lines are the boundaries of the present experiment ($\phi = 0.5°$ and $2.8°$) with $E_i = 102$ meV, calculated from eq. (1). The dashed line is the upper boundary accessible with a spectrometer having the lowest scattering angle of $\phi = 5°$.

The spin-wave dispersion curves in $La_{0.8}Sr_{0.2}MnO_3$ observed at $T = 6$ and 245 K were well fitted to $E_p(Q) = DQ^2$ with $D = 130 \pm 4$ and 88 ± 2 meVÅ^2, respectively. In previous works measuring spin waves using single crystals, an isotropic quadratic dispersion curve with $D =$ 131 meVÅ^2 at $T = 14$ K and 89 meVÅ^2 at 250 K for $La_{0.8}Sr_{0.2}MnO_3$ was reported [5, 6]. At T = 14 K and $Q < 0.3$ Å^{-1} from an appropriate reciprocal point, the dispersion curve along [001] was well fitted to $E_p(Q) = DQ^2$ with $D = 131$ meVÅ^2 and was identical to that along [111] [6].

The spin-wave dispersion curve of $SrRuO_3$ observed at $T = 7$ K was well fitted to a quadratic form with an apparent energy gap: $E_p(Q) = E_0 + DQ^2$ with $E_0 = 2.2 \pm 0.3$ meV and $D = 48 \pm 5$ meVÅ^2, by using the data points for $Q = 0.15 - 0.3$ Å^{-1}. Spin waves in $La_{0.8}Sr_{0.2}MnO_3$ show the isotropic dispersion relations in smaller Q range, however the curves gradually shift with each other for $Q > 0.3$ Å^{-1} due to the crystalline effect. We applied this experimental evidence to the case of $SrRuO_3$: the dispersion relation at $Q \leq 0.3$ Å^{-1} may follow a form of $E_0 + DQ^2$.

4. Permanent magnet $Nd_2Fe_{14}{}^{11}B$

$Nd_2Fe_{14}B$ is a well-known song permanent magnet with $T_C = 580$ K and the saturation magnetization of 1.6 T. At room temperature, all spins are aligned along the c*-axis. Below $T = 130$ K the system exhibits a spin reorientation. Because the unit cell includes four chemical formulae of $Nd_2Fe_{14}B$ in a tetragonal lattice, spin-wave branches are expected to be complicated. In a previous inelastic neutron scattering experiment using a single-crystalline sample of $Nd_2Fe_{14}{}^{11}B$, where ^{11}B was used for minimizing the neutron absorption, a spin-wave branch was detected only along the c*-axis around (002) at $T = 6$ and 295 K [8]. We performed NBS experiments in a $Nd_2Fe_{14}{}^{11}B$ polycrystalline sample at $T = 6$ and 300 K with $E_i = 257$ meV and $\Delta E = 5.7$ meV. The sample volume was approximately 5 cm^3. The spin-wave peaks in the observed energy spectra at $T = 300$ K were well fitted to Gaussian scattering functions with widths determined from the resolution including the dispersion slope, and the peak positions were determined in Fig. 2. The observed peak positions were on the dispersion curve along the

c*-axis reported in the previous experiment. No magnetic peaks were observed at $T = 6$ K, this result is also consistent with the dispersion curve along the c*-axis at $T = 6$ K reported in the previous experiment.

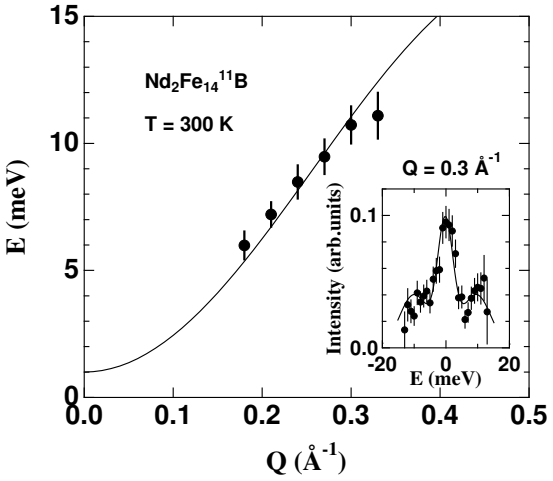

Figure 2. Spin-wave peak positions for $Nd_2Fe_{14}{}^{11}B$. The solid line is a dispersion curve along the c*-axis determined using a single crystal in ref. [8]. The inset shows a typical excitation spectrum with a fitted curve.

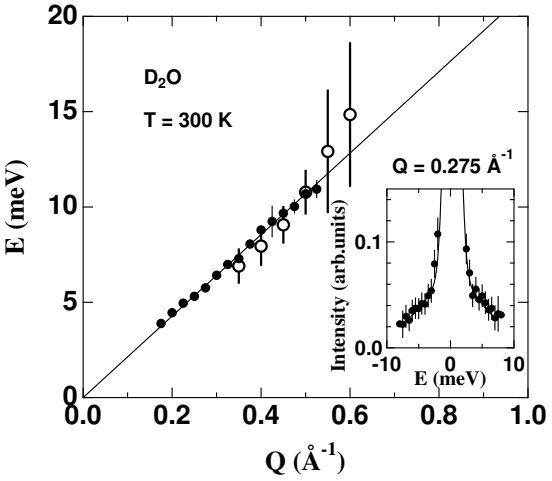

Figure 3. Dispersion relation for phononic excitations, the fast sound, in D_2O (closed circles). The open circles are previous results in ref. [9]. The solid line is a fitted line. The inset shows a typical excitation spectrum with a fitted curve.

5. Liquid D_2O

Phononic excitations in a liquid D_2O were measured at $T = 300$ K with $E_i = 102$ meV and $\Delta E = 2.0$ meV. The sample volume was approximately 15 cm^3. The background-subtracted spectra showed excitation peaks and a resolution-limited elastic peak. The excitation peaks were fitted with a damped harmonic oscillator scattering function convoluted with the resolution width multiplied by a temperature factor and the peak positions $E_p(Q)$ were determined. The present analysis was identical to that for the previous inelastic neutron scattering experiment [9]. As shown in Fig. 3, the Q dependence of the observed peak positions down to $Q = 0.02$ Å^{-1} was well fitted to $E_p(Q) = cQ$ with $c = 21.4 \pm 0.2$ meVÅ, which is equivalent to $c = 3250 \pm 30$ m/s. The observed sound velocity c was in good agreement with that for the fast sound observed in the previous experiment with $E_i = 80$ meV and $\Delta E = 4.8$ meV down to $Q = 0.035$ Å^{-1} [9].

6. Summary

We demonstrated the feasibility of NBS experiments on the HRC by observing coherent excitations in $La_{0.8}Sr_{0.2}MnO_3$, $SrRuO_3$, $Nd_2Fe_{14}{}^{11}B$ polycrystals and a liquid D_2O. In the current research in material sciences, most of the newly synthesized materials are chemically and structurally complicated such that large single crystals are not always possible to be synthesized. NBS experiments on the HRC bring about new opportunities to such material development using polycrystalline and powder samples, because the HRC at the pulsed neutron source facilitates the possibility of extending the (Q, E) space using the sub-eV range of incident neutron energies.

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