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Spin dynamics of Er^{3+} in $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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Neutron scattering techniques are used to study the magnetic excitations of the Er ions in superconducting $\text{ErBa}_2\text{Cu}_3\text{O}_7$, a system where the Er ions order antiferromagnetically at $T_N=0.62$ K and the sublattice magnetization obeys the relation for a 2D $S=1/2$ Ising model. Our data, which were taken on a cold neutron triple axis spectrometer using a horizontally focusing analyzer, reveal that there is a large gap of about 0.20 meV in the excitation spectrum, about four times the thermal energy kT_N , as expected for an Ising system. However, we also observe dispersion of the spin waves in the a - b plane. These excitations are found to renormalize with temperature and only diffuse paramagnetic scattering is observed well above the ordering temperature. © 1997 American Institute of Physics. [S0021-8979(97)16108-4]

The rare earth (R) ions in $\text{RBa}_2\text{Cu}_3\text{O}_{6+x}$ ($0 < x < 1$) exhibit very interesting ordering behavior such as two-dimensional (2D) and 3D long range order, and short range correlations.¹⁻⁷ The R ions occupy the centered position in a pseudotetragonal cell ($a \approx b$ and $c \approx 3a$), between the two Cu-O plane layers. There is no direct overlap of orbitals of nearest neighbor R ions, which are separated by a distance a , and therefore the magnetic interactions are very weak. Magnetic interactions along the c direction are much weaker than in the a - b plane because the nearest-neighbor distance is about three times longer along the c direction than in the a - b plane, and this anisotropy leads to a 2D magnetic behavior in this system.³ The Er ions in $\text{ErBa}_2\text{Cu}_3\text{O}_7$ order antiferromagnetically at $T_N=0.62$ K with the moments aligned ferromagnetically along the b direction and antiferromagnetically along a and c , and the sublattice magnetization is found to follow the relation for a 2D $S=1/2$ Ising model very well.^{2,4} Reduction in the oxygen concentration strongly affects the magnetic ordering and, in particular, no long range ordering is observed in $\text{ErBa}_2\text{Cu}_3\text{O}_6$. Despite these interesting magnetic properties, the spin wave excitations of the rare earth ions in any of the layered cuprate systems have not been measured previously, for two reasons. One is the unavailability of large high-quality single crystals, and the other is the need for high experimental energy resolution in order to observe these low energy excitations.

Recently, we were able to grow a 1.5 g single crystal, and with a cold neutron triple axis spectrometer, we have been able to observe the spin wave excitations in a reasonable counting time. We were also able to take advantage of a horizontally focusing analyzer, which can coarsen the in-plane Q resolution without loss of energy resolution. This is particularly advantageous for low dimensional systems, where the resolution can be relaxed in the nondispersive direction, increasing the signal without sacrificing information. Our data along the (1,1,0) direction show a large gap of about 0.2 meV, but with a significant amount of dispersion, while no dispersion is observed along the c direction. These observations are in good agreement with the behavior expected for a 2D Ising system.

A 1.5 g single crystal of $\text{ErBa}_2\text{Cu}_3\text{O}_7$ was grown by a seeding technique of the undercooled melt at a constant temperature.⁸ The crystal was oxygenated at 400 °C for 7 days to obtain fully oxygenated composition of $\text{ErBa}_2\text{Cu}_3\text{O}_7$. For the low temperature measurements, a pumped ^3He cryostat with a low temperature capability of 0.3 K was used. Neutron diffraction measurements to study the magnetic order of Er, and preliminary inelastic neutron scattering experiments, were conducted on the BT-2 triple axis spectrometer with pyrolytic graphite (PG) monochromator and analyzer at the Research Reactor at the National Institute of Standards and Technology (NIST). Incident energies of 14.8 and 5.0 meV were employed. Inelastic measurements with cold neutrons were conducted on the NG-5 spin polarized inelastic neutron scattering (SPINS) spectrometer at the Cold Neutron Research Facility (CNRF) at NIST. A PG(002) vertically focusing monochromator and a horizontally focusing analyzer⁹ were used in these measurements, with a Be filter to suppress the higher-order wavelength contaminations. No collimators were used for the cold neutron experiments. Some measurements were made in the $(h,0,l)$ scattering plane, but twinning in the sample complicates the analysis of those inelastic data. However, twinning does not significantly affect measurements in the (1,1,0) direction, and therefore most of the inelastic experiments reported here were conducted in the (h,h,l) scattering plane. We selected a fixed final energy of 2.6 meV to obtain good energy resolution while still being able to take advantage of the horizontally focusing analyzer.

Our neutron diffraction measurements showed that the Er ions order antiferromagnetically at $T_N=0.62$ K in this crystal, with the moments aligned ferromagnetically along the b direction and antiferromagnetically along a and c , in agreement with the previous studies.^{2,4} The temperature dependence of magnetic intensities showed 2D Ising behavior with a sharp transition at T_N . Since this behavior is not expected for the oxygen deficient compound,⁴ we believe our sample is fully oxygenated.

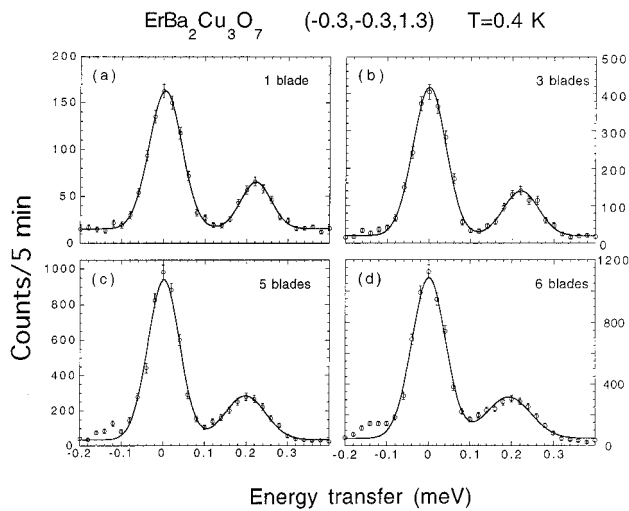


FIG. 1. Neutron scattering intensity with (a) one blade, (b) three blades, (c) five blades, and (d) six blades of the horizontally focusing analyzer, as a function of energy transfer at 0.4 K. These data were collected at $(-0.3, -0.3, 1.3)$ with a final energy of 2.6 meV. Solid curves are Gaussian fits to the data.

Inelastic neutron scattering data with 5 meV incident energy on the BT-2 spectrometer indicated the existence of magnetic excitations. However, the intensities were weak, and this prevented us from fully measuring the dispersion relation. Hence it was important to increase the observable magnetic intensity, and this can be achieved by using a cold neutron spectrometer. The cold source triple axis spectrometer has two advantages over the thermal neutron instruments. One is that experimental energies as low as 2.4 meV can be employed, and this will provide improved energy resolution. The second obvious advantage is that the flux is about 5 times higher on the cold source at this energy.

We also used a horizontally focusing analyzer⁹ to get higher magnetic intensity. The intention of using this analyzer is to increase the horizontal acceptance of the analyzer system for neutrons scattered from the sample. The focusing analyzer consists of eleven 2-cm-wide PG(002) blades. Each of these blades can be rotated independently, and in addition the entire assembly can be rotated such that the scattered neutrons are focused onto the detector. This horizontal focusing technique increases the signal by coarsening the momentum Q resolution, and this focusing can be done without affecting the energy resolution. Inelastic data taken with a fixed final energy of 2.6 meV, with one single flat blade, are shown in Fig. 1(a). These measurements were taken at $Q = (-0.3, -0.3, 1.3)$. The energy resolution in this configuration is about 0.1 meV (full width at half-maximum). These data show an elastic peak, and an inelastic peak at 0.221(3) meV. This inelastic peak originates from magnetic excitations. Data taken with three blades in the focusing mode are shown in Fig. 1(b). These data clearly show that compared to the single-blade case, both the elastic and inelastic peak intensities increase, without affecting the energy resolution.

Although the horizontally focusing analyzer increases the signal, it also increases the probability of collecting unwanted intensities onto the detector due to the high acceptance angle and lack of collimators in the experiment, and

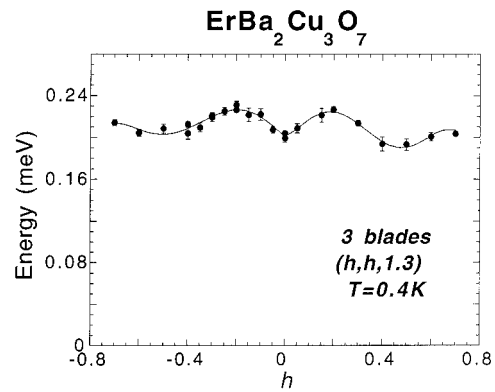


FIG. 2. The dispersion of magnetic excitations along $(1,1,0)$ direction. The solid curve is a guide to the eye. There is a large gap in the excitation spectrum of about 0.2 meV, but with significant dispersion.

this may make it difficult to correctly identify the magnetic excitations that are of interest. This effect can be easily noticed in the data taken with five and six blades [Figs. 1(c) and 1(d)]. These figures show some extra intensity around -0.12 and $+0.15$ meV that does not originate from magnetic excitations. In order to avoid this problem, we conducted the rest of the experiment with only three blades.

We have measured the dispersion along the $(1,1,0)$ direction, and the data are shown in Fig. 2. These data indicate that there is a substantial gap of about 0.2 meV, equivalent to a thermal energy of 2.3 K, about $3.75 T_N$. This large gap is expected for an Ising system. However, for an ideal Ising system, we expect a dispersionless spin wave mode, while we observe a significant dispersion along the $(1,1,0)$ direction. The spin wave energy, which is 0.20 meV at $(0,0,1.3)$, gradually increases to 0.23 meV at $(\pm 0.2, \pm 0.2, 1.3)$ and then decreases again. Our measurements along the c axis indicate that these excitations show no measurable dispersion in that direction, and this behavior is expected because the magnetic interactions are very weak along the c direction.

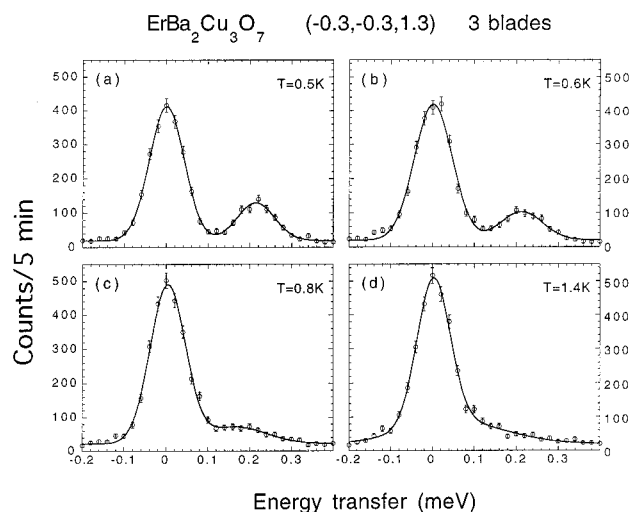


FIG. 3. The neutron scattering intensity as a function of energy transfer at (a) 0.5, (b) 0.6, (c) 0.8, and (d) 1.0 K. Solid curves are fits to the data.

The temperature dependence of the inelastic spectrum is shown in Fig. 3, and were taken with the same conditions as that of Fig. 1(b). With increasing temperature, the peaks that arise from spin wave excitations become broad, and then become purely diffusive well above T_N (0.62 K). Detailed measurements of the critical scattering and the field dependence of the inelastic scattering are in progress.

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