Low Energy Phonons and Triplet-Phonon Coupling in the Singlet Ground State of $SrCu_2(BO_3)_2$

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Two-dimensional (2D) copper-oxides, in which conducting holes are introduced, provide the framework for much exotic behavior, with high temperature superconductivity being the best appreciated example. While there are many instances of quasi 1D spin ladder or spin chain compounds, there are somewhat few good quasi 2D experimental compounds. Strontium copper borate $SrCu_2(BO_3)_2$ is a unique quantum spin system among the known quantum magnets, with 2D arrangement of spin-1/2 Cu dimers that has attracted much interest in recent years. Its ground state is known to be a collective singlet state with a single-triplet gap of ~ 3 meV and two-triplet gap of ~ 4.8 meV [1,2]. This compound is close to a quantum critical point (QCP) and the ground state evolves from a gapped singlet to a gapless long-range antiferromagnetic state as a function of the magnetic interactions [3]. Additionally, 2D magnetic systems are of particular importance because of their relevance to the study of high temperature superconductivity. Shastry and Kumar argue the Mott-Hubbard gap will be suppressed by doping into a Shastry-Sutherland system, while several superconducting phases are predicted at low temperature [4,5]. Much interest has also focused on magnetization plateaus which appear [6] beyond 20 T in $SrCu_2(BO_3)_2$. Strong magnetic fields generate triplets within a background of singlets, which can undergo Bose-Einstein condensation (BEC) at densities determined by the applied magnetic field.

The studies on $SrCu_2(BO_3)_2$ material have been mostly focused on the magnetic properties of the system. However, recently there has been increasing interest in the possible role of the spin-lattice effects in $SrCu_2(BO_3)_2$. It has been suggested that there is a relation between spin-lattice interaction in $SrCu_2(BO_3)_2$ and the magnetic dynamics at low temperatures and high magnetic fields [3]. Such a strong interplay between crystal and magnetic properties is well known and one of the characteristic aspects of low dimensional spin systems. In $SrCu_2(BO_3)_2$ a lattice distortion is thought to stabilize the spin superstructure associated with the 1/8 magnetization plateau, the first observation of magnetization plateau in a quasi 2D material [1,5]. In general, lattice distortions in $SrCu_2(BO_3)_2$ are thought to allow magnetic interactions which are otherwise forbidden in a more symmetric environment by lowering the crystal symmetry [7]. For example, buckling of the $CuBO_3$ planes allows components of Dzyaloshinskii-Moriya interactions. These subleading Dzyaloshinskii-Moriya interactions weakly split the three triplet modes even in zero applied magnetic field [7-9]. For this reason an analysis of the vibrational modes in this compound is of interest. Recent infrared spectroscopy measurements with polarized light on $SrCu_2(BO_3)_2$ have shown a phonon excitation peak in the *a-b* plane at 55 meV which splits below 15 K [10,11]. This indicates that the singlet ground state is likely associated with the spin-phonon coupling. The temperature scale is consistent with the decrease in the dc-susceptibility, characteristic of a gapped spin system.

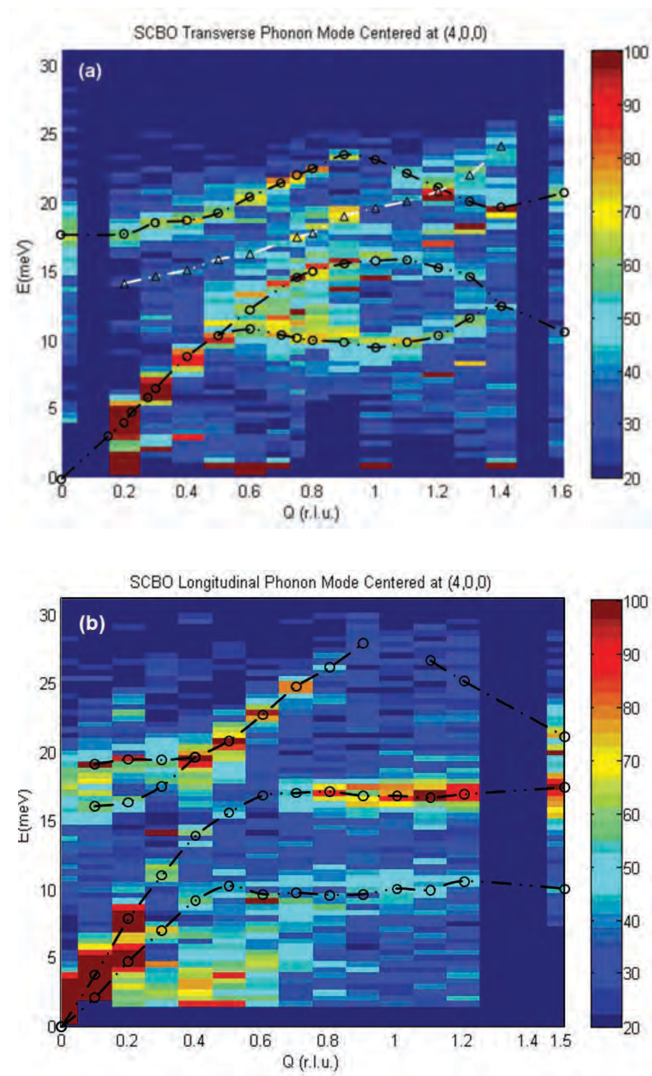

Fig. 1 Two sets of measurements were performed near the zone centre **Q** = (4,0,0,) and T~ 150 K. (a) Transverse dispersion curve for acoustic modes and low energy transverse optical mode. (b) Longitudinal dispersion curve for acoustic modes along with the low energy longitudinal optical modes.

These results intrigued us to use inelastic neutron scattering technique to study the acoustic and optical phonons in the single crystal of $SrCu_2(BO_3)_2$. In this experiment, we investigated the phonons propagating within the *a-b* basel plane using C5 triple-axis-spectrometer. The crystal was mounted in a closed cycle displex with its (H, K, 0) plane coincident with the horizontal plane, such that wavevectors near the **Q** = (4, 0, 0) zone centre could be accessed. These measurements employed pyrolitic graphite as both monochromator (vertically focused) and analyzer (flat) crystals with a fixed final neutron energy of 3.52 THz. Two pyrolitic graphite filters were placed in the scattered beam to reduce higher order contamination. Soller slits determined the horizontal collimation equal to [none, 0.48°, 0.55°,1.2°], using the convention [source-monochromator, monochromator-sample, sample-analyzer, analyzer-detector]. We initially performed a survey of the low energy lattice dynamics in this system. Two sets of measurements were performed near the zone centre **Q** = (4, 0, 0) to study transverse low energy and longitudinal low energy modes. The results are shown in figure 1. The dispersion curves of the transverse and longitudinal acoustic phonons at low Q and low energy transfers along with the low energy optical phonon modes are clearly observed in this figure. The dispersion curves are in agreement with the zone boundary condition of this systems, which in the (H, K, 0) scattering plane is: h + k = 2n.

In addition we investigated the influence of the triplet and multi-triplet bound state magnetic excitations on the lattice vibrations. For this part of the experiment we used a collimation setting of [none, 0.273°, 0.477°, 1.2°] achieving an energy resolution of ~ 0.5 meV. We investigated closely the phonon dispersion curve centered at **Q** = (4,0,0) in the vicinity of ~ 0.725 THz (3 meV) where the triplet magnetic excitation

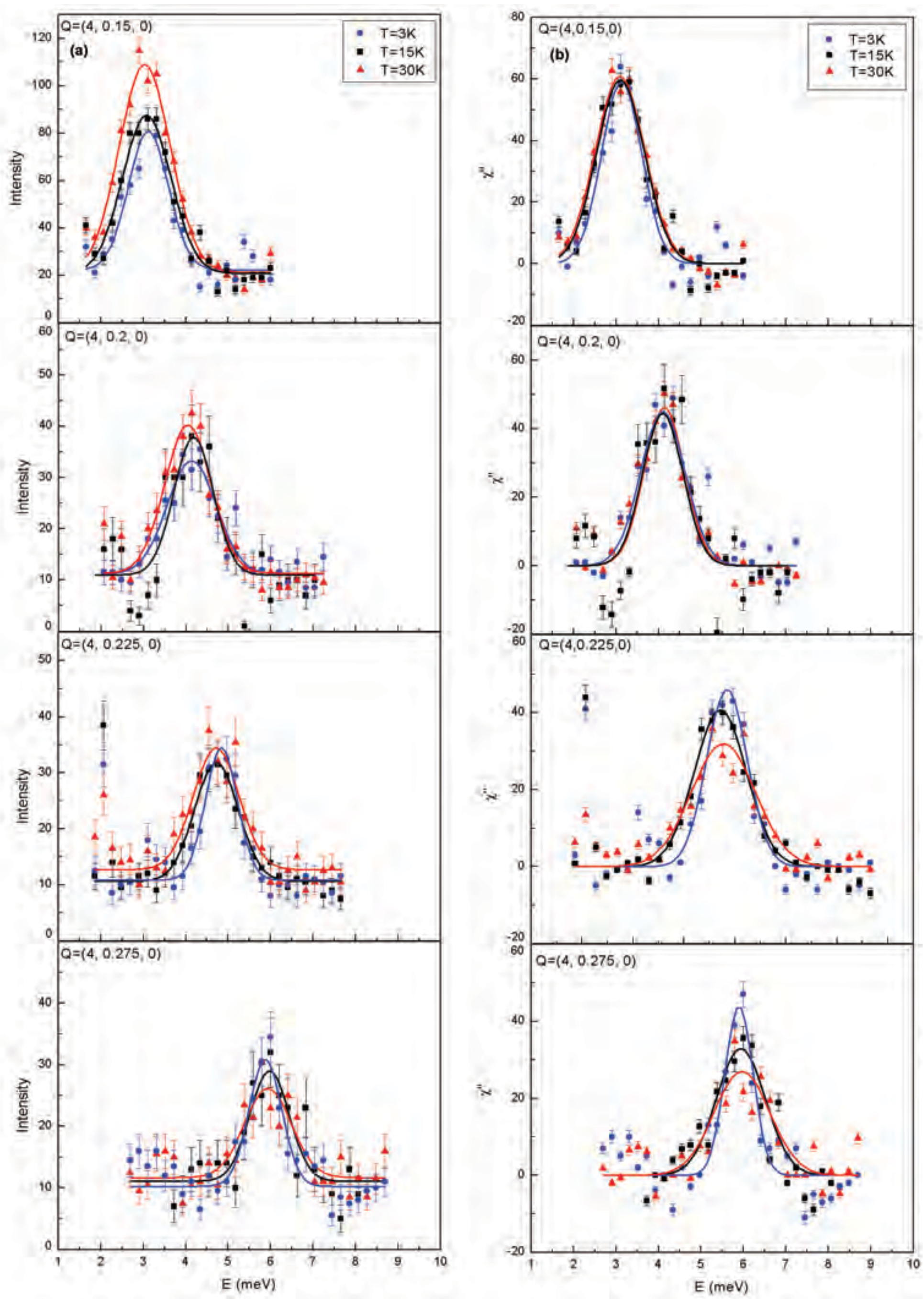

Fig 2: The results of constant Q scans for low energy transverse phonons centered at **Q** = (4, 0, 0) Bragg peak in $SrCu_2(BO_3)_2$. (a) As observed inelastic neutron scattering data and (b) corrected for background and Bose factor. Solid lines are resolution convoluted fits to the data as described in the text.

crosses the acoustic phonon branches. We also studied the inelastic scattering in the energy range near ~ 1.185 THz (4.9 meV) where the two-triplet bound state magnetic resonance exists. We measured the low-energy phonons as a function of temperature below and above the temperature at which the collective singlet ground state is fully established (~ 8 K). This allowed us to investigate the influence of the singlet-triplet and multi-triplet bound state magnetic excitations that only exist within the singlet ground state on the lattice vibrations.

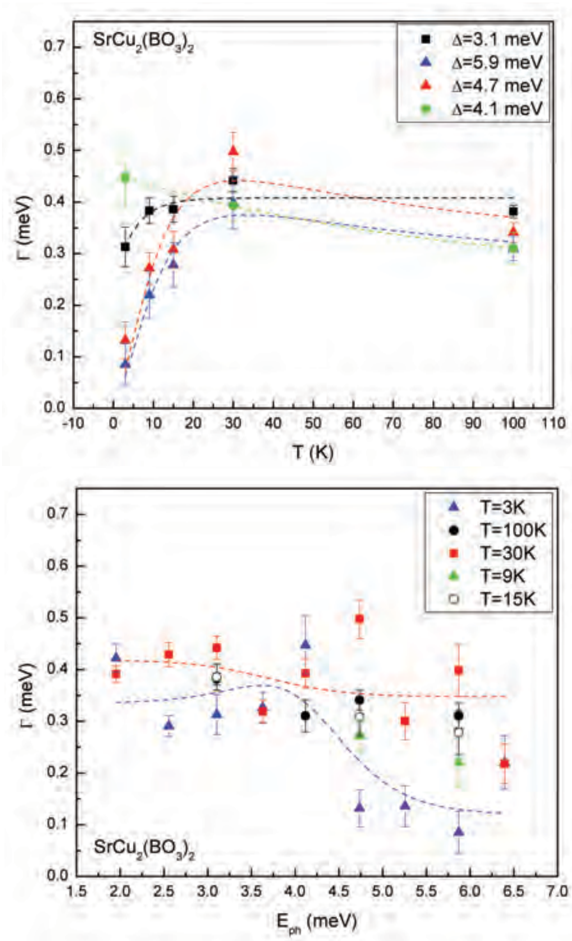

Fig 3: *The extracted phonon excitations widths (Γ), proportional to the phonon inverse lifetime, are shown as a function of (a) temperature and (b) phonon energy. It is clear that only the high energy, low temperature phonons have a resolution limited width. The results show that there is evidence for coupling between phonon degrees of freedom and at least two-triplet spin excitation. Dashed lines are guide to the eye.*

Figure 2 shows the results of constant Q scans for low energy transverse phonons centered at **Q** = (4, 0, 0) Bragg peak in $SrCu_2(BO_3)_2$. The results of raw inelastic neutron scattering data are shown in figure 2(a) whereas figure 2(b) shows the results of inelastic neutron scattering data corrected for background and Bose factor so as to isolate the dynamic susceptibility for transverse acoustic phonons in $SrCu_2(BO_3)_2$. As seen from this figure, there is a significant enhancement of the imaginary part of the dynamic susceptibility associated with the transverse acoustic phonons as they cross the two triplet bound state at an energy transfer of ~ 4.8 meV. It is interesting that this enhancement occurs as the phonons cross the two triplet bound state, as opposed to the energy of one triplet spin excitations.

Figure 2 shows representative data with accompanying fits used to extract the lifetime of phonons. The data was fit to damped harmonic oscillator model, appropriately convoluted with the instrumental resolution function. Resolution convoluted fits of the data are shown as the solid lines in figure 2, as seen the fits describe the data well. Such fits allow us to extract the inverse of triplet excitation lifetime (Γ), shown in Figure 3 as a function of temperature and phonon energy. Surprisingly only phonons at higher energies close to multi-triplet bound states and low temperatures where there is a singlet ground state have a resolution limited line width. As seen from Figure 3b, the phonon lifetime stays short at lower energies for all temperatures.

While further experimental and theoretical work is required to fully understand this behaviour, our results clearly indicate a significant coupling between the singlet ground state and the low lying phonons in $SrCu_2(BO_3)_2$ specifically at higher energies where the multi-triplet excitation exists.

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