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Multiple singlet and triplet states in the 2D orthogonal-dimer compound $SrCu_2(BO_3)_2$

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Abstract

The compound $SrCu_2(BO_3)_2$ is a quantum spin system with a 2D arrangement of orthogonal spin dimers, an exact singlet ground state and a singlet–triplet gap $\Delta_{01} = 34$ K. Triplet excitations in this compound are extremely localized and strongly interacting due to the strong inherent frustration. Experimentally, two triplet modes were observed both in neutron scattering and ESR investigations. In Raman scattering experiments further well-defined modes with energies close to Δ_{01} exist for low temperatures ($T \ll \Delta_{01}$). The properties of these modes resemble the magnetic bound states found in dimerized spin chain and spin ladder systems.

Keywords: $SrCu_2(BO_3)_2$; Spin gap; Bound states; Raman scattering

1. Introduction

Low-dimensional quantum spin systems with a finite spin gap have been a subject of intense research in the last years. The singlet ground state is realized in one dimension in the dimerized phase of spin-Peierls systems ($T < T_{SP}$), alternating spin chains or spin ladders. Compounds that have been investigated within this context are, e.g., $CuGeO_3$, α'-NaV_2O_5, $(VO)_2P_2O_7$ or $Sr_{14-x}Ca_xCu_{24}O_{41}$. Of even greater interest is the existence of a gap in a two-dimensional (2D) spin system. Unfortunately only very few 2D spin systems, such as CaV_4O_9, are discussed in which the gap is relatively large, i.e. not caused by spin anisotropies. Recently, $SrCu_2(BO_3)_2$ with a layered structure has been identified as a 2D system [1,2]. Furthermore, due to its exceptional exchange topology this compound can be regarded as a model system with an exactly known ground state given by the direct product of singlet pairs [3,4].

$SrCu_2(BO_3)_2$ has a tetragonal unit cell (D_{2d}) with Cu^{2+} ions that carry spin $s = \frac{1}{2}$ and have distances to nearest and next-nearest Cu neighbors of approximately 2.9 and 5.1 Å, respectively. A spin dimer consists of a neighboring pair of planar rectangular CuO_4. Dimers are connected orthogonally by a triangular BO_3. This 2D lattice in the *ab* plane of the compound is topologically equivalent to the Shastry–Sutherland lattice studied earlier [3] and is given by a 2D square lattice with an additional alternating diagonal interaction. This interaction serves as a spin frustration. The Shastry–Sutherland lattice thus can be considered as a 2D analog of the Majumdar–Ghosh model for a one-dimensional zigzag spin chain [5]. The strength of the next-nearest-neighbor or interdimer interaction $J_2 = 68$ K, which brings a frustration into the system, and an intradimer coupling of $J_1 = 100$ K have been estimated [4]. It has been highlighted that the ratio $J_2/J_1 = 0.68$ is just below the critical value of $J_2/J_1 = 0.70$ separating the exact dimer state and a Néel-ordered state. Thus this compound is very close to a quantum critical point between a gapfull exact dimer and a gapless phase [4,6].

Recent ESR [7] and neutron scattering investigations [8] observed triplet excitations from the singlet ground state fixing the spin gap at $\Delta_{01} = 34$ K. This triplet

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branch observed at 3 meV has a very small dispersion of only 0.2 meV. This value is much smaller than expected theoretically [6] and points to extremely localized triplet excitations. In addition, a second triplet branch $\Delta'_{01} = 55\,\mathrm{K} \equiv 4.7\,\mathrm{meV}$ with a larger dispersion is observed. A further interesting aspect of this compound is its magnetic phase diagram. Quantized plateaus at $\frac{1}{4}$ and $\frac{1}{8}$ of the saturated Cu moment are observed in the magnetization for fields up to 50 T [9]. Although similar effects are well known for 1D spin systems this is the first observation of ordered triplet phases in a 2D compound [4].

2. Results and discussion

Raman scattering experiments on $SrCu_2(BO_3)_2$ at room temperature show numerous phonon lines in the frequency range 30–1200 cm^{-1}. In the low-energy region drastic changes happen for $T < 5$ K leading to the appearance of several new modes. In Fig. 1 Raman spectra are shown at $T = 1.7$ K and in several scattering configurations within the *ab* plane of the crystal. Spectra in the upper panel show data in $(b'b')$: $A_1 + B_2$ and (aa): $A_1 + B_1$ configuration, while the lower panel gives $(a'b')$: B_1 and (ab): B_2. Symmetry components are denoted with respect to the D_{2d} point group using $b' = a + b$.

We first concentrate on the (ab) component in the lower panel. Modes with weak intensity are observed at 24.5, 31 and 40 cm^{-1}. All other signals in this polarization are extremely weak and probably have their origin in spurious contributions from other polarizations. The first and the third mode are identical in frequency with the triplet modes observed in ESR and neutron scattering discussed above. In the $(a'b')$ polarization additional modes are observed at 29.3 and 56 cm^{-1}. These modes show up with an intensity three times larger than that of the modes in (ab) polarization. They are also observed in the (aa) and $(b'b')$ polarizations given in the upper panel. Further broadened modes with no distinct selection rules appear at 45–47 and 65–75 cm^{-1}. In contrast, the mode at 61 cm^{-1} observed both in (aa) and $(b'b')$ polarization has a well-defined A_1 symmetry, a smaller line shape and exists also at higher temperatures. All these properties are in favor of its interpretation as a phonon mode.

The low-excitation spectrum of $SrCu_2(BO_3)_2$ is very detailed and complex. However, comparing these observations with earlier experiments on 1D spin-gap systems [10,11] several conclusions are drawn. The temperature dependence of the intensity and the energy of the above modes, except the 61 cm^{-1} mode, both point to their origin in the singlet–triplet gap. Selection rules allow to divide the observed modes into three groups as triplet excitations (B_2 symmetry, three modes), predominant singlet excitations ($A_1 + B_1$ symmetry, two modes) and

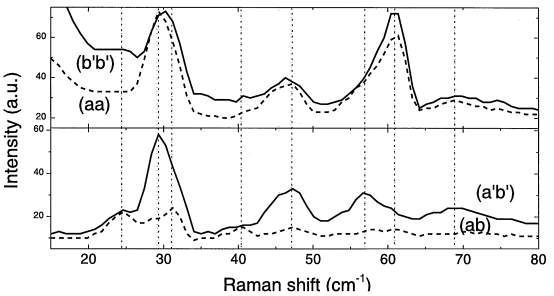

Fig. 1. Low-energy Raman spectra of $SrCu_2(BO_3)_2$ in different scattering geometries in the *ab* plane (see text) at $T = 1.7$ K. The vertical dotted lines indicate the frequencies of the observed modes. With the exception of the phonon mode at 61 cm^{-1} all other modes are of magnetic origin.

mixed modes (two modes). Indeed, in systems with strongly localized triplets a number of singlet and triplet bound states are possible. We propose to identify $\Delta_{01} = 24.5$ cm^{-1} as the original triplet branch and $\Delta'_{01} = 40$ cm^{-1} as the first triplet bound state. The modes at 29.3, and 56 cm^{-1} may be assigned to singlet bound states. All other modes have to be attributed to higher-order bound states as they are either very weak in intensity or triplet states are not observed in neutron and ESR experiments [12].

The binding energy of the first triplet (first singlet) bound state with respect to $2\Delta_{01}$ is $E_t = 9$ cm^{-1} ($E_s = 19.7$ cm^{-1}). These binding energies are large if compared to 1D frustrated spin systems. However, due to the strong localization [4] of the triplet excitations on the orthogonal dimer sites strong binding effects are expected. Evidence for a multitude of magnetic excitations is also given in recent ESR experiments [7]. In conclusion, Raman scattering experiments at low temperatures on $SrCu_2(BO_3)_2$ showed a multitude of magnetic excitations with energies close to the singlet–triplet gap. The number, selection rules and other properties of these modes are more complicated compared with 1D alternating chain systems.

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