Optical spectroscopy of \((\text{La, Ca})_{14}\text{Cu}_{24}\text{O}_{41}\) spin ladders: comparison of experiment and theory

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Abstract

Transmission and reflectivity of \(\text{La}_x\text{Ca}_{14-x}\text{Cu}_{24}\text{O}_{41}\) two-leg spin-1/2 ladders were measured in the mid-infrared regime between 500 and 12,000 cm\(^{-1}\). This allows us to determine the optical conductivity \(\sigma_1\) directly and with high sensitivity. Here we show data for \(x=4\) and 5 with the electrical field polarized parallel to the rungs (\(E||a\)) and to the legs (\(E||c\)). Three characteristic peaks are identified as magnetic excitations by comparison with two different theoretical calculations.

Key words:
spin ladder, bound state, phonon-assisted 2-magnon absorption

The quantum nature of magnetic excitations in spin-1/2 systems and in particular the role of quantum fluctuations in low dimensions are a fascinating subject. Antiferromagnetic (AF) \(S=1/2\) Heisenberg ladders represent an intermediate step between one-dimensional (1D) chains and the 2D \(\text{CuO}_2\) layers of undoped high-\(T_c\) superconductors. The elementary excitations of the ladders can be described as triplets or as interacting spinons. Topics of current interest are theoretical predictions of 2-triplet bound states [1], the size of the exchange coupling along the rungs (\(J_\perp\)) and the legs (\(J_\parallel\)) as well as the role of the ring exchange \(J_{\text{cyc}}\) [2]. We address these issues in \(\text{La}_x\text{Ca}_{14-x}\text{Cu}_{24}\text{O}_{41}\) which contains layers with \(\text{Cu}_2\text{O}_3\) two-leg AF \(S=1/2\) ladders [3].

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A La content of $x=6$ corresponds to nominally undoped samples, i.e. Cu$^{2+}$, but single phase crystals were obtained only for $x \lesssim 5$ [4]. Reflectivity and transmission data for $x=5$ and 4 at 4 K are plotted in Fig. 1 along with the deduced real part $\sigma_1$ of the optical conductivity. Except for the strong phonon signature at low frequencies the reflectivity is featureless, demonstrating that reflectivity measurements with subsequent Kramers–Kronig transformation are not adequate to resolve small values of $\sigma_1$. The transmission, however, is much more sensitive to weak absorption and combining transmission and reflectivity one can determine $\sigma_1$ most accurately.

The spectra can be divided into 3 different regimes. Below $\approx 1300 \text{ cm}^{-1}$ the rise of $\sigma_1$ is due to phonon absorption. The high frequency behavior is dominated by an electronic background that increases with hole doping, i.e. decreasing $x$. To analyze the peaks in the intermediate region we subtracted this background using an exponential fit (thin lines in Fig. 1). After subtraction the remaining features are almost independent of $x$ (see Fig. 2 for $x=5$). We interpret these excitations in terms of phonon-assisted two-magnon absorption [5] which has been used to describe $\sigma_1$ of the undoped 2D cuprates (e.g. YBa$_2$Cu$_3$O$_6$ [6]) and of the 1D S=$1/2$ chain Sr$_2$CuO$_3$ [7]. Due to spin conservation two magnons are excited. The simultaneous excitation of a phonon provides the symmetry breaking necessary to bypass the selection rule and it guarantees momentum conservation [3,5,6]. Since the exchange coupling in the chains is $\approx 2$ orders of magnitude smaller than in the ladders, we attribute the observed absorption to the ladders.

In Fig. 2 we compare the magnetic contribution to $\sigma_1$ of the lowest nominal doping $x=5$ (dashed lines; one hole per formula unit) with 2 different theoretical calculations. One approach is related to 1D spinon physics and describes the spins in terms of Jordan–Wigner fermions with a long-ranged phase factor (thin lines in Fig. 2). The other approach starts from isolated singlets on each rung, i.e. $J_{\parallel}=0$, with local triplet excitations. Using continuous unitary transformations [8], finite $J_{\parallel}$ is then treated as a perturbation that creates delocalized, dressed triplets (thick lines in Fig. 2). Concerning the dispersion of the elementary excitation (triplet or “magnon”), the differences between both theories are $\lesssim 10$–20% [3]. Both show a dispersing two-triplet bound state with $S_{\text{tot}}=0$ that leaves the two-triplet continuum at $k \gtrsim 0.3\pi$. The maximum of this bound state at $k \approx \pi/2$ and its minimum at $k=\pi$ yield van-Hove singularities in the density of states that cause the 2 peaks at 2800 and 2140 cm$^{-1}$, respectively. Both theories are in excellent agreement with the experimental data for $J_{\parallel}/J_1 \approx 1$–1.2 with $J_1 \approx 1020$–1100 cm$^{-1}$. Further confirmation of our interpretation is the reduced spectral weight of the peak at 2140 cm$^{-1}$ for $E_{||} > a$ caused by a selection rule arising from symmetry [3]. We have thus verified the theoretical predictions of a two-triplet bound state [1]. Finally, the broad peak at around 4000 cm$^{-1}$ is identified with the two-triplet continuum.
A ratio of $J_\parallel/J_\perp \approx 1$ seems to be in conflict with several former results of other techniques, proposing $J_\parallel/J_\perp \gtrsim 1.5$ (see discussion in [2]). Such large values can be excluded on the basis of our results [3]. The introduction of a ring exchange $J_{\text{cyc}} \approx 0.15 J_\parallel$ resolves this issue in favor of $J_\parallel/J_\perp \approx 1–1.1$ [9].

In conclusion, the existence of a two-triplet bound state is verified in the two-leg $S=1/2$ ladders of La$_x$Ca$_{14-x}$Cu$_{24}$O$_{41}$ ($x=5$ and 4). We obtain the values of the exchange constants $J_\parallel \approx 1020–1100$ cm$^{-1}$ and $J_\parallel/J_\perp \approx 1–1.2$.

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References

[1] G.S. Uhrig and H.J. Schulz, Phys. Rev. B 54, R9624 (1996); erratum ibid. 58, 2900 (1998); O.P. Sushkov and V.N. Kotov, Phys. Rev. Lett. 81, 1941 (1998); S. Trebst et al., Phys. Rev. Lett. 85, 4373 (2000); C. Jurecka and W. Brenig, Phys. Rev. B 61, 14307 (2000).

[2] D.C. Johnston et al., cond-mat/0001147.

[3] M. Windt et al., Phys. Rev. Lett. (2001), in press.

[4] U. Ammerahl and A. Revcolevschi, J. Crystal Growth 197, 825 (1999); U. Ammerahl, PhD thesis, Univ. of Cologne, 2000.

[5] J. Lorenzana and G.A. Sawatzky, Phys. Rev. Lett. 74, 1867 (1995); Phys. Rev. B 52, 9576 (1995).

[6] M. Grüninger et al., Phys. Rev. B 62, 12422 (2000).

[7] H. Suzuura et al., Phys. Rev. Lett. 76, 2579 (1996); J. Lorenzana and R. Eder, Phys. Rev. B 55, R3358 (1997).

[8] C. Knetter and G.S. Uhrig, Eur. Phys. J B 13, 209 (2000); C. Knetter et al., Phys. Rev. Lett. (2001), in press.

[9] M. Matsuda et al., Phys. Rev. B 62, 8903 (2000); J. Appl. Phys. 87, 6271 (2000).
Fig. 1. Reflectivity, transmission and optical conductivity of $\text{La}_{x}\text{Ca}_{14-x}\text{Cu}_{24}\text{O}_{41}$. Solid lines: $x=4$; dashed: $x=5$; thin lines in lower panel: exponential fits to the electronic background. Transmission sample thicknesses: $\approx 60 \, \mu\text{m} \,(\approx 44 \, \mu\text{m})$ for $x=4$ (5).

Fig. 2. Magnetic contribution to $\sigma_1$ of $\text{La}_5\text{Ca}_9\text{Cu}_{24}\text{O}_{41}$ (dashed lines) compared with calculations using optimized perturbation (thick lines, $J_\perp = J_\parallel = 1020 \, \text{cm}^{-1}$) and Jordan-Wigner fermions (thin lines, $1100 \, \text{cm}^{-1}$), respectively. The assumed phonon energy is $600 \, \text{cm}^{-1}$. An exponential electronic background was subtracted from the measured data of Fig. 1. Note the small values of $\sigma_1 \leq 3 \, (\Omega\text{cm})^{-1}$. 