ELSEVIER

Physica B 312–313 (2002) 617–618

PHYSICA B

www.elsevier.com/locate/physb

Optical spectroscopy of $(La, Ca)_{14}Cu_{24}O_{41}$ spin ladders: comparison of experiment and theory

M. Grüninger[a,*], M. Windt[a], T. Nunner[b], C. Knetter[c], K.P. Schmidt[c], G.S. Uhrig[c], T. Kopp[b], A. Freimuth[a], U. Ammerahl[a,d], B. Büchner[a,e], A. Revcolevschi[d]

[a] *II. Physikalisches Institut, Universität zu Köln, Zülpicherstr. 77, 50937 Köln, Germany*
[b] *Experimentalphysik VI, Universität Augsburg, 86135 Augsburg, Germany*
[c] *Institut für Theoretische Physik, Universität zu Köln, 50937 Köln, Germany*
[d] *Laboratoire de Physico-chimie, Université Paris-Sud, 91405 Orsay, France*
[e] *II. Physikalisches Institut, RWTH-Aachen, 52056 Aachen, Germany*

Abstract

Transmission and reflectivity of $La_xCa_{14-x}Cu_{24}O_{41}$ two-leg spin-$\frac{1}{2}$ ladders were measured in the mid-infrared regime between 500 and 12 000 cm^{-1}. This allows us to determine the optical conductivity σ_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.

PACS: 74.72.Jt; 75.10.Jm; 75.40.Gb; 78.30.−j

Keywords: Spin ladder; Bound state; Phonon-assisted 2-magnon absorption

The quantum nature of magnetic excitations in spin-$\frac{1}{2}$ systems and in particular the role of quantum fluctuations in low dimensions are a fascinating subject. Antiferromagnetic (AF) $S = \frac{1}{2}$ Heisenberg ladders represent an intermediate step between one-dimensional (1D) chains and the 2D 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_{cyc} [2]. We address these issues in $La_xCa_{14-x}Cu_{24}O_{41}$ which contains layers with Cu_2O_3 two-leg AF $S = \frac{1}{2}$ ladders [3].

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 σ_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 σ_1. The transmission, however, is much more sensitive to weak absorption and combining transmission and reflectivity one can determine σ_1 most accurately.

The spectra can be divided into 3 different regimes. Below $\approx 1300\ cm^{-1}$ the rise of σ_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 (dotted 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 σ_1 of the undoped 2D cuprates (e.g. $YBa_2Cu_3O_6$ [6]) and of the 1D $S = \frac{1}{2}$ chain Sr_2CuO_3 [7]. Due to spin conservation two magnons are excited. The simultaneous excitation of a phonon provides the symmetry breaking necessary to bypass

*Corresponding author. Tel.: +49-221-470-3595; fax: +49-221-470-6708.

E-mail address: grueninger@ph2.uni-koeln.de (M. Grüninger).

0921-4526/02/$ - see front matter
PII: S 0 9 2 1 - 4 5 2 6 (0 1) 0 1 1 9 9 - 1

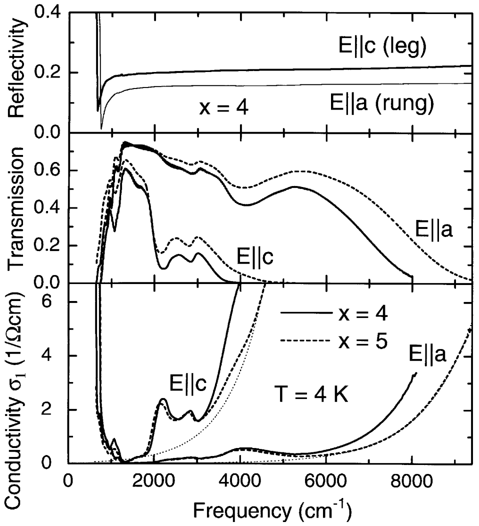

Fig. 1. Reflectivity, transmission and optical conductivity of $La_xCa_{14-x}Cu_{24}O_{41}$. Solid lines: $x = 4$; dashed: $x = 5$; dotted lines in lower panel: exponential fits to the electronic background. Transmission sample thicknesses: ≈ 60 μm (≈ 44 μm) for $x = 4$ (5).

the selection rule and it guarantees momentum conservation [3,5,6]. Since the exchange coupling in the chains is ≈ 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 σ_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 (dash-dot 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 (solid 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 $S_{tot} = 0$ 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 $J_{\parallel}/J_{\perp} \approx 1$–1.2 with $J_{\parallel} \approx 1020$–1100 cm^{-1}. Further confirmation of our interpretation is the reduced spectral weight of the peak at 2140 cm^{-1} for $E \parallel a$ 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

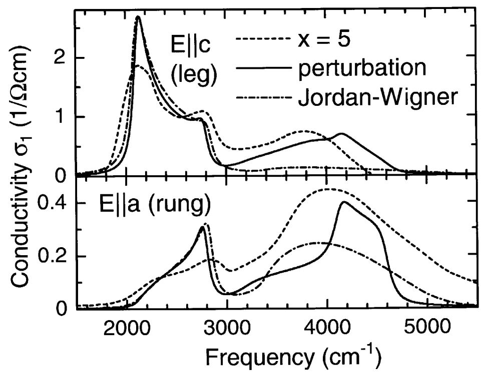

Fig. 2. Magnetic contribution to σ_1 of $La_5Ca_9Cu_{24}O_{41}$ (dashed lines) compared with calculations using optimized perturbation (solid lines, $J_{\perp} = J_{\parallel} = 1020$ cm^{-1}) and Jordan–Wigner fermions (dash-dot lines, 1100 cm^{-1}), respectively. The assumed phonon energy is 600 cm^{-1}. An exponential electronic background was subtracted from the measured data of Fig. 1. Note the small values of $\sigma_1 \leqslant 3$ Ω cm^{-1}.

$J_{\parallel}/J_{\perp} \gtrsim 1.5$ (see discussion in Ref. [2]). Such large values can be excluded on the basis of our results [3]. The introduction of a ring exchange $J_{cyc} \approx 0.15 J_{\parallel}$ 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 $S = \frac{1}{2}$ ladders of $La_xCa_{14-x}Cu_{24}O_{41}$ ($x = 5$ 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.

This project is supported by the DFG (FR 754/2-1, SP 1073 and SFB 484), by the BMBF (13N6918/1) and by the DAAD within the scope of PROCOPE.

References

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

[2] D.C. Johnston, M. Troyer, S. Miyahara, D. Lidsky, K. Ueda, M. Azuma, Z. Hiroi, M. Takano, M. Isobe, Y. Ueda, M.A. Korotin, V.I. Anisimov, A.V. Mahajan, L.L. Miller, cond-mat/0001147.

[3] M. Windt, et al., Phys. Rev. Lett. 87 (2001) 127002.

[4] U. Ammerahl, A. Revcolevschi, J. Crystal Growth 197 (1999) 825;
U. Ammerahl, Ph.D. Thesis, University of Cologne, 2000.

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

[6] M. Grüninger, D. van der Marel, A. Damascelli, A. Erb, T. Nunner, T. Kopp, Phys. Rev. B 62 (2000) 12422.

[7] H. Suzuura, H. Yasuhara, A. Furusaki, N. Nagaosa, Y. Tokura, Phys. Rev. Lett. 76 (1996) 2579;
J. Lorenzana, R. Eder, Phys. Rev. B 55 (1997) R3358.

[8] C. Knetter, G.S. Uhrig, Eur. Phys. J B 13 (2000) 209;
C. Knetter, et al., Phys. Rev. Lett. 87 (2001) 167204.

[9] M. Matsuda, K. Katsumata, R.S. Eccleston, S. Brehmer, H.-J. Mikeska, Phys. Rev. B 62 (2000) 8903;
M. Matsuda, et al., J. Appl. Phys. 87 (2000) 6271.