PHONON-ASSISTED MAGNETIC ABSORPTION OF $(La,Ca)_{14}Cu_{24}O_{41}$: CONTRIBUTION OF DIFFERENT PHONON MODES*

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We obtain the phonon spectrum for a simplified structure $(La,Ca)_2Cu_2O_3$ using a shell model calculation. The external photon–electron–phonon vertex is expanded up to fifth order in the copper–oxygen hopping (t_{pd}) and oxygen–oxygen hopping (t_{pp}) and the resulting form factors for the phonon-assisted magnetic contribution to the optical conductivity $\sigma(\omega)$ of $(La,Ca)_{14}Cu_{24}O_{41}$ are analyzed. In this way we can understand the shift of approx. 60 cm^{-1} observed experimentally between the upper two-triplet bound states observed in $\sigma_{\mathrm{rung}}(\omega)$ and $\sigma_{\mathrm{leg}}(\omega)$.

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Optical spectroscopy constitutes a valuable tool for the investigation of low-dimensional quantum antiferromagnets. Measurements of the optical conductivity $\sigma(\omega)$ of the spin ladder compound $(La,Ca)_{14}Cu_{24}O_{41}$ [1] have established the first experimental verification for the existence of the two-triplet $S = 0$ bound states. Two peaks at $2140\,\mathrm{cm}^{-1}$ and $2780\,\mathrm{cm}^{-1}$ correspond to the $S = 0$ bound states with momentum $p_x = \pi$ and $p_x = \pi/2$, respectively, where the momentum is provided by the simultaneous excitation of a phonon. In a recent publication [2] we have shown that a thorough analysis of $\sigma(\omega)$ of $(La,Ca)_{14}Cu_{24}O_{41}$ allows to derive a minimal model, which is the basis for all theoretical investigations of these spin-ladders systems.

In order to collect further evidence in favor of our minimal model we use the dynamical DMRG [3] to calculate the entire spectrum of $\sigma(\omega)$ for an $N = 80$ site ladder. We obtain excellent agreement with the experimental result of $La_{5.2}Ca_{8.8}Cu_{24}O_{41}$ (see Fig. 1) using an anisotropy of $J/J_\perp = 1.3$

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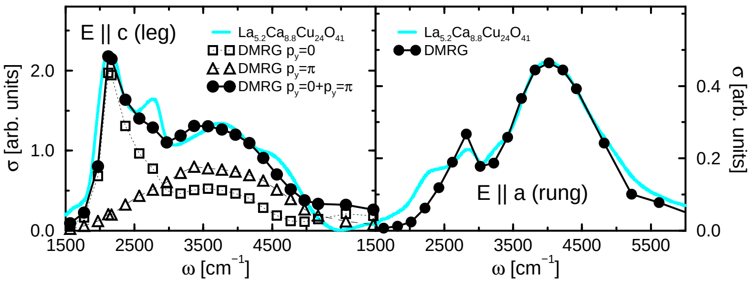

Fig. 1. Symbols: Correction-vector results for the optical conductivity $\sigma(\omega)$ of an $N = 80$ site ladder with polarization of the electrical field E along the legs (left panel) and along the rungs (right panel), using $J/J_\perp = 1.3$, $J_{\rm cyc}/J_\perp = 0.2$, $J_\perp = 1000\,{\rm cm}^{-1}$, $\omega_{ph}^{\rm leg} = 570\,{\rm cm}^{-1}$, $\omega_{ph}^{\rm rung} = 620\,{\rm cm}^{-1}$ and a finite broadening of $\delta = 0.1J_\perp$. The leg polarization contains two contributions, where the two legs are in-phase ($p_y = 0$) or out-of-phase ($p_y = \pi$) with each other. Gray line: magnetic contribution to $\sigma(\omega)$ of $La_{5.2}Ca_{8.8}Cu_{24}O_{41}$ at $T = 4\,{\rm K}$ after subtraction of an electronic background [2].

for the exchange couplings along the legs and rungs and a cyclic spin exchange of $J_{\rm cyc}/J_\perp = 0.2$ with $J_\perp = 1000$ cm^{-1} [2]. In spite of the overall impressive agreement with the experimental line shape, there still remain some minor inconsistencies which will be discussed here: *(i)* Experimentally, the frequency of the upper bound state of 2780 cm^{-1} is approx. 60 cm^{-1} lower in $\sigma_{\rm leg}(\omega)$ than in $\sigma_{\rm rung}(\omega)$. *(ii)* In $\sigma_{\rm leg}(\omega)$ the upper bound state (corresponding to $p_x \approx \pi/2$) is suppressed much stronger in the DMRG calculation than in the experimental spectrum.

In order to address these questions it is necessary to consider the phonons and the process, which couples light to the magnetic excitations in more detail. To obtain some information about the frequencies and the displacement patterns of the phonon modes of (La,Ca)$_{14}$Cu$_{24}$O$_{41}$, we use a shell model calculation with the parameters listed in Ref. [5]. We believe that the incommensurate CuO_2 chains have only little influence on the oxygen vibrations with polarization along the Cu_2O_3-planes, whereas it is important to include the (La,Ca)-ions to obtain the correct values of the oxygen bending modes. The dispersion of the relevant oxygen modes and corresponding displacement patterns at (0,0) as obtained for the simplified structure (La,Ca)$_2$Cu$_2$O$_3$ are displayed in Fig. 2 and Fig. 3, respectively.

This knowledge about the phonon modes in (La,Ca)$_{14}$Cu$_{24}$O$_{41}$ is important because the excitation of a phonon [4] is necessary to break the inversion symmetry between two neighboring copper spins and thus to generate a finite dipole moment for a coupled "phonon–two triplet" excitation

$$\sigma(\omega) \sim -\omega \sum_{p_x} \sum_{p_y=0,\pi} |f_p|^2 \, {\rm Im}\langle\langle \delta B_{-p}; \delta B_p \rangle\rangle_{(\omega-\omega_{ph})}\,, \qquad (1)$$

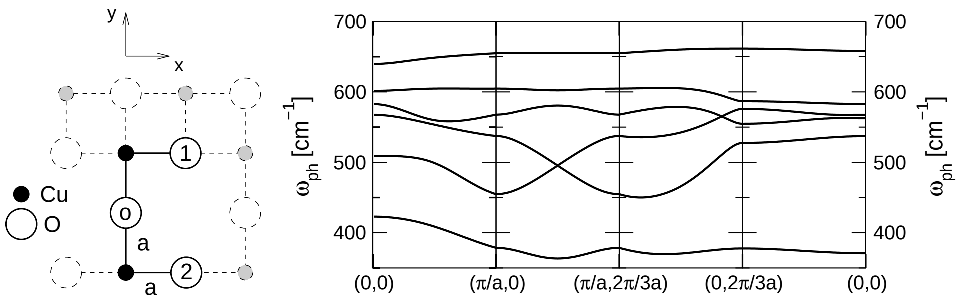

Fig. 2. Oxygen phonon modes with polarization within the Cu_2O_3-planes calculated with the shell model parameters of Ref. [5].

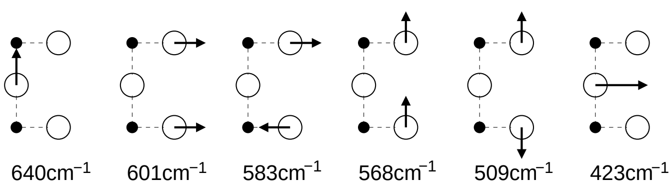

Fig. 3. Displacement patterns and frequencies at (0,0).

where the spin-flip operators $\delta B_p^{\text{rung}} = \frac{1}{N}\sum_i e^{ipr_i}\left(\boldsymbol{S}_{i,l}\boldsymbol{S}_{i,r} - \langle \boldsymbol{S}_{i,l}\boldsymbol{S}_{i,r}\rangle\right)$ and $\delta B_p^{\text{leg}} = \frac{1}{N}\sum_i\sum_{k=l,r} e^{ipr_{i,k}}\left(\boldsymbol{S}_{i,k}\boldsymbol{S}_{i+1,k} - \langle \boldsymbol{S}_{i,k}\boldsymbol{S}_{i+1,k}\rangle\right)$ correspond to polarization of the electrical field along the legs and the rungs, respectively. The vertex function f_p depends on the phonon mode involved and can be derived in a perturbation expansion of the super exchange coupling J in the electric field and in the displacements of the oxygen ions. Expanding f_p to the lowest two orders (t_{pd}^4 and $t_{pd}^4 t_{pp}$) using a set of typical parameters for the 3-band Hubbard model in the cuprates ($U_d = 8.8\,\text{eV}$, $U_p = 6\,\text{eV}$, $t_{pd} = 1.3\,\text{eV}$, $t_{pp} = 0.65\,\text{eV}$, $\Delta = 3.5\,\text{eV}$) we obtain the following form factors for a phonon with wave-vector parallel to the legs (*i.e.* $p_y^{\text{ph}} = 0$)

- polarization || leg, in-phase excitation of the legs:

$$\begin{aligned} f_p^{\text{leg,ip}} &= (0.7J^{(4)} - 0.1J^{(5)} + (1.7J^{(4)} + 2.6J^{(5)})\sin^2\frac{p_x a}{2})(u_{1x} + u_{2x}) \\ &- \ 0.3J^{(5)}\cos\frac{p_x a}{2}(2u_{0x} + u_{1x} + u_{2x}) \\ &+ \ i(0.8J^{(4)} + 2.6J^{(5)})\sin\frac{p_x a}{2}(u_{1y} - u_{2y}) \end{aligned} \tag{2}$$

- polarization || leg, out-of-phase excitation of the legs:

$$\begin{aligned} f_p^{\text{leg,op}} &= \left((1.7J^{(4)} + 2.6J^{(5)})\sin^2\frac{p_x a}{2} + 0.3J^{(5)}\cos\frac{p_x a}{2}\right)(u_{1x} - u_{2x}) \\ &+ \ i\,(0.8J^{(4)} + 2.6J^{(5)})\sin\frac{p_x a}{2}(2u_{0y} - u_{1y} - u_{2y}) \end{aligned} \tag{3}$$

- polarization || rung:

$$f_p^{\text{rung}} = (1.5J^{(4)}+1.2J^{(5)})u_{0y} - \Big(0.4J^{(4)}+0.7J^{(5)}+0.3J^{(5)}\cos\frac{p_x a}{2}\Big) \times (u_{1y}+u_{2y}) i\,(0.8J^{(4)}+2.6J^{(5)})\sin\frac{p_x a}{2}(u_{2x}-u_{1x}) \qquad (4)$$

with $J^{(4)} = 0.250\,\text{eV}$ and $J^{(5)} = 0.569\,\text{eV}$ for the 4th and 5th order contributions to the super exchange coupling J without electric field and phonons.

On this basis we are able to discuss the two questions put up in the introduction. For *(i)* we compare the frequencies of the phonon modes which give the strongest contribution to $\sigma(\omega)$. For $\sigma_{\text{rung}}(\omega)$ this is a stretching mode of the rung oxygen (see the term $\propto u_{0y}$ in Eq. (4)) which has a frequency of $\omega_1 = 640\,\text{cm}^{-1}$ and is the highest mode in Fig. 2. The largest contribution to the in-phase part of $\sigma_{\text{leg}}(\omega)$, which contains the bound states, is caused by the in-phase stretching ("$u_{1x}+u_{2x}$") and by the out-of-phase bending ("$u_{1y}-u_{2y}$") mode of the leg-oxygens (see Eq. (2)). In our shell-model calculation these two modes (see Fig. 2 and Fig. 3) have a frequency of $\omega_2 = 601\,\text{cm}^{-1}$ and $\omega_4 = 568\,\text{cm}^{-1}$, respectively. This corresponds to a difference of about ω_1-$\omega_2 = 39\text{cm}^{-1}$ and $\omega_1 - \omega_4 = 72\,\text{cm}^{-1}$ between the frequency of the upper bound state in $\sigma_{\text{rung}}(\omega)$ and $\sigma_{\text{leg}}(\omega)$, which is in good agreement with the experimentally observed shift of approx. $60\,\text{cm}^{-1}$.

Question *(ii)* is related to the form factors used in Eq. (1). The upper bound state (corresponding to $p_x \approx \pi/2$) is strongly suppressed in $\sigma_{\text{leg}}(\omega)$ due to a form factor $|f_p|^2 \sim \sin^4\frac{p_x a}{2}$, which is the dominant term to order $t_{pd}^4 \propto J^{(4)}$ and has been used in Fig. 1. In the next order, *i.e.* $t_{pd}^4 t_{pp} \propto J^{(5)}$, however, a form factor $|f_p|^2 \sim \sin^2\frac{p_x a}{2}$ gains similar strength as can be inhered from Eq. (2). The inclusion of this prefactor will pronounce the upper bound state with respect to the lower bound state.

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REFERENCES

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

[2] T.S. Nunner *et al.*, *Phys. Rev.* **B66**, 180404(R) (2002).

[3] T.D. Kühner, S.R. White, *Phys. Rev.* **B60**, 335 (1999).

[4] J. Lorenzana, G.A. Sawatzky, *Phys. Rev. Lett.* **74**, 1867 (1995); *Phys. Rev.* **B52**, 9576 (1995).

[5] V.N. Popov, *J. Phys. Condens. Matter* **7**, 1625 (1995); M.V. Abrashev, A.P. Litvinchuck, C. Thomsen, V.N. Popov, *Phys. Rev.* **B55**, 9136 (1997).