Q-dependence of spin excitations in high-T_C cuprates from spin–phonon coupling

2009 J. Phys.: Condens. Matter 21 142202

(http://iopscience.iop.org/0953-8984/21/14/142202)

IOP FTC
IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER
J. Phys.: Condens. Matter **21** (2009) 142202 (4pp) doi:10.1088/0953-8984/21/14/142202

FAST TRACK COMMUNICATION

Q-dependence of spin excitations in high-T_C cuprates from spin–phonon coupling

T Jarlborg

Département de Physique de la Matière Condensée, University of Geneva,
24 Quai Ernest-Ansermet, CH-1211 Geneva 4, Switzerland

Received 23 January 2009
Published 5 March 2009
Online at stacks.iop.org/JPhysCM/21/142202

Abstract
An understanding of spin excitations in cuprates is essential since the mechanism of high-T_C superconductivity might be linked to spin fluctuations. Band calculations for 'one-dimensional' unit cells of La_2CuO_4 show larger coupling (spin–phonon coupling, SPC) between anti-ferromagnetic spin waves and O-phonons than for Cu- or La-phonons. When this result is applied to a two-dimensional, free-electron like band, it leads to an 'hourglass' shape of the spin excitation spectrum, as in recent experiments. Isotope shifts and doping dependences of the excitations are discussed.

Many properties of high-T_C copper oxides, including the mechanism behind superconductivity, are not understood, but there is a consensus about the existence and the importance of complex superstructures and pseudogaps. Spatial, stripe-like regions with excitations of charge and spin modulations were detected quite early from neutron scattering experiments [1]. Angular resolved photoemission established that the bandstructures agree well with calculations, where one dispersive band gives rise to an ubiquitous barrel-like Fermi-surface (FS) (or an FS-'arc' when displayed in 1/4th of the Brillouin zone) [2, 3]. However, this arc is truncated and survives only in the diagonal direction ($k_x \approx k_y$) at low temperature, T, [4]. Phonons enter also into this complexity, as shown by the softening of some phonon branches at certain dopings [5–7], by isotope effects on the pseudogap near the temperature T^*, on T_C [8], and on the gap structure itself [9]. Non-commensurate, q-dependent spin excitations appear as side spots in neutron scattering at (0.5-q, 0.5) and (0.5, 0.5-q), where q varies linearly as a function of doping, x, up to a saturation at $x \approx 0.12$ [10]. Recent measurements have established that the underlying spin excitations have a characteristic 'hourglass' shaped ($\bar{q}, \omega$)-dispersion, even at non-superconducting compositions, and different explanations have been discussed [11–13].

Interaction between phonons and electronic states have been suggested for the cuprates [14, 15], and *ab initio* band calculations show large spin–phonon coupling (SPC) within the CuO plane [16]. This means a stronger anti-ferromagnetic (AFM) wave when it coexists with a phonon [17]. The *ab initio* band results, in combination with two-dimensional (2D) free-electron like bands, have been used for modeling of many normal state properties of the high-T_C materials [18, 19]. Phonon softening, dynamic stripes, correlation between $\bar{q}$ and x, smearing of the non-diagonal part of the FS, and abrupt disappearance of the spin fluctuations at a certain T^*, are possible consequences of SPC within a rather conventional band. Here, from calculations for different types of phonon distortions, it is proposed that also the characteristic spin dispersion can be understood in terms of SPC.

First, we discuss the *ab initio* band calculations made for $La_{(2-x)}Ba_xCuO_4$ (LBCO), where the virtual crystal approximation (VCA) is applied to La sites to account for the doping. The calculations are made using the linear muffin-tin orbital method (LMTO) in the local spin-density approximation (LSDA), as has been described previously [18]. Phonon distortion amplitudes (u) and the size of Cu moments (m) in spin waves are necessary input to these calculations. The T-dependences $u^2 \approx 3k_BT/K_u$ and $m^2 \approx k_BT/K_m$ are valid for not too low T. Here $K_p = d^2E/dp^2$, E is the total energy, and $p = u$ or m, respectively [18]. The force constants K_u for the different atoms are taken from measurements on $YBa_2Cu_3O_7$ [20, 21]. The resulting u/a_0, where a_0 is the lattice constant, shown in table 1 for $T \approx 100$ K (near T_C and T^*), compare well with experiment [22]. An approximate

Table 1. Three first lines: phonon distortion amplitudes, u/a_0, induced potential shifts, V_q^p (mRyd), and exchange splitting, V_q^m (mRyd), caused by spin waves ($\mu_B H = \pm 5$ mRyd). All shifts/splittings refer to the maximal value on a Cu site, and they are determined in self-consistent LMTO calculations for supercells of length $8a_0$. Remaining lines: the partial characters of the phonon DOS, N_i, for different sites, i, as estimated from [23]. pl-O means plane oxygen and ap-O means apical oxygen.

Wave	No-phonon	pl-O_x	La_z	ap-O_z	Cu_x
u/a_0	—	0.014	0.021	0.017	0.024
V_q^p	—	15	2	5	3.5
V_q^m	8	12.5	12	10	8.5
$N_{\text{pl-O}}$	—	0.6	0.0	0.2	0.2
N_{La}	—	0.0	0.65	0.0	0.35
$N_{\text{ap-O}}$	—	0.35	0.0	0.65	0.0
N_{Cu}	—	0.2	0.2	0.0	0.6

calculation of K_m for a short wave in $HgBa_2CuO_4$ (HBCO) corresponds to magnetic moments m on Cu of about $\sim 0.09\ \mu_B$ at 100 K [17] for an applied magnetic field of ± 5 mRyd [18]. The calculations consider distortions (and coexisting AFM spin waves) in cells extending $8a_0$ along $\bar{x}$, with displacements of La and apical O along $\bar{z}$, and Cu and planar O along $\bar{x}$. For the latter phonon there is a positive SPC when the nodes of the AFM waves are located at the 'compressed' Cu sites (when the O atoms move towards the Cu). Optimal SPC for displacements of out-of-plane atoms (La and apical O) occurs when these atoms move towards the CuO plane near the region of the AFM node. (A modulation of apical O with the opposite phase will make the spin wave slightly weaker than a spin wave without a phonon.) The calculated results for the maximal potential shifts on Cu caused by phonons (V_q^p) and by spin waves (V_q^m in the spin polarized potential) for the four types of movements in the cell are shown in the table, together with the result for V_q^m without phonons.

Phonon energies and partial phonon density-of-states (DOS) for each site, N_{site} calculated by Chen and Callaway [23] for Nd_2CuO_4, can be assumed to be representative for LBCO. The main La-modes are at 10–20 meV, Cu at 20–30 meV, planar O near 50 ± 20 meV, and apical O at 60 ± 15 meV [20, 23]. The estimations of N_{site} from [23] are shown in the last part of table 1. The LMTO results calculated for waves of length $8a_0$ are rescaled to be valid for the longer waves at doping $x = 0.16$ (12–$13a_0$ in the model calculation), when V_q^p and V_q^m are larger by factors ~ 1.1 and ~ 1.5, respectively, see [19]. The total V_q^t becomes

$$V_q^t = \sum_i (1.1 V_{q,i}^p + 1.5 V_{q,i}^m) N_i, \qquad (1)$$

where i is the site index. The resulting V_q^t are 17, 18, 23 and 22 mRyd at the energies centered around 15 (La), 25 (Cu), 50 (plane O) and 60 meV (apical O), respectively. As discussed later, spin waves at higher energy should be independent of phonons, and $V_q^p = 0$.

In a second step, we use the parameters V_q^t in a 2D nearly free-electron (NFE) model. The AFM spin arrangement on neighboring Cu along [1, 0, 0] in undoped LBCO corresponds to a potential perturbation, $V(\bar{x}) = V_q^t \exp(-i\bar{Q} \cdot \bar{x})$ (and equivalently with $\bar{y}$ along [0, 1, 0]). The periodicity in real space is defined through the Cu–Cu distance, and a gap of size $2V_q^t$ appears at the zone boundary, at $\bar{Q}/2$. A further modulation ($\bar{q}$) of this order into one-dimensional (1D) stripes perpendicular to $\bar{x}$ (or 'checkerboards' in two dimensions along $\bar{x}$ and $\bar{y}$) is achieved by a multiplication of the potential by $\exp(i\bar{q} \cdot \bar{x})$, where $\bar{q} < \bar{Q}$. In total, this makes $V(\bar{x}) = V_q^t \exp(-i\bar{Q}_x \cdot \bar{x})$, where $\bar{Q}_x = \bar{Q} - \bar{q}$. The periodicity of this potential is now larger, and the gap moves away from $\bar{Q}/2$ to $(\bar{Q} - \bar{q})/2$. Magnetic side spots appear at $\bar{q}/2$ surrounding $\bar{Q}/2$ in probes which can separate the two periodicities. A 3×3 eigenvalue problem with matrix elements $H_{11} = E - k_x^2 - k_y^2$, $H_{22} = E - (k_x - Q_x)^2 - k_y^2$, $H_{33} = E - k_x^2 - (k_y - Q_y)^2$, $H_{12} = H_{13} = V_q^t$ and $H_{23} = 0$, is solved. The lowest FE band (when $V_q^t = 0$) contains two electrons up to $E_F \sim 0.15$ Ryd, when the effective mass is one. The total band width (and the x- and y-dispersion) is quite close to that of the real band, and the NFE band is sufficient to demonstrate the effects of SPC. The model is entirely 2D, which is reasonable in view of the very small band dispersion along $\bar{k}_z$ of real band structures for the cuprates [16].

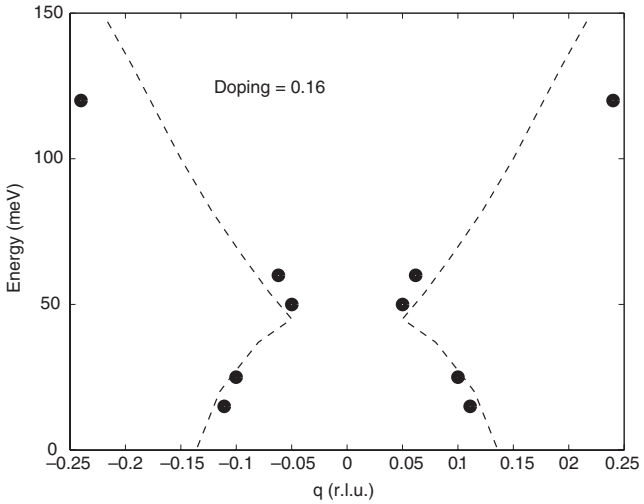

Figure 1. Filled circles: calculated $q - \hbar\omega$ relation from the 2D-NFE model and the parameters V_q^t for doping $x = 0.16$. The solution without SPC at the largest energy is not very precise (see text). Broken line: approximate shape of the experimental dispersion as it is read from figure 3c in the work of Vignolle *et al* [13] for $La_{2-x}Sr_xCuO_4$ at $x = 0.16$. The q-value appears to be smaller (by ~ 0.01 reciprocal lattice units (r.l.u.) at low energy and by ~ 0.04 at high energy) in measurements on LBCO at lower doping, $x = 0.125$ [12].

An important result of the 2D-NFE model is that it leads to a correlation between doping and the amplitude of V_q^t [19]. The reason is that the gap opens along $(k_x, 0)$ and $(0, k_y)$, but not in the diagonal direction. The combined effect is that the dip in the total DOS (at which E_F should fall for optimal doping) will not appear at the same band filling for a small and a wide gap, even if the q-vector is the same. Alternatively, since the gap should appear at the same energy (at E_F) for different V_q^t, one has to search for the q-vector that fits with E_F for each V_q^t. The present calculations are made for 0.16 holes per Cu (which is close to x in [13]) with V_q^t from equation (1).

The results are shown in figure 1 together with experimental data from [13]. The spectrum is shaped like an

hourglass with a 'waist' at intermediate energy, since the SPC strength differs with energy (no SPC would make $\vec{q}$ equal for all energies). The points below 70 meV are for the coupling to the four types of phonons. The general shape of the $q-\omega$-dependence and the amplitudes of q are similar to those in [13], with the smallest q at about 0.05 for SPC dominated by O-p-phonons around 50 meV. The weaker SPC for La at low energy makes q larger. A larger band mass leads to smaller amplitudes of q. If no mixing of the phonon modes (through the N_{site}-coefficients) were made it would increase the variations with q, but the general hourglass shape remains.

Spin waves and phonons are tied together at the same frequency and q-vector for the optimal mechanism of SPC. (Short-lived spin fluctuations at high energy could somehow profit from an interaction with slow phonon distortions, but stronger damping makes them probably less important.) Therefore, spin waves with higher frequency than the phonons cannot profit from the normal SPC, whereby $V_q^p = 0$. The energy of this 'phonon independent' result is put rather arbitrarily at ~120 meV in figure 1, which is about twice the highest phonon frequency. The solution without SPC leads to a wave which is close to (or slightly shorter than) the cell in the LMTO calculation, so the factor 1.5 in equation (1), is reduced to ~1.0 for this (phonon free) mode. Thus, the appropriate value of V_q^t is 8 mRyd, with the solution at $q = 0.24$, see figure 1. In contrast to the cases with SPC and larger V_q^t, this solution implies a very short wave, 4–5a_0 only, where V_q^t might be reduced even more. This will, because of the self-consistent feedback between spin density and potential, lead towards a vanishing spin wave [18]. Therefore, it is difficult to imagine larger q for spin excitations with small V_q^t at this doping.

Other high-energy solutions with small V_q^t exist for non-equal q-vectors along x and y [19], where the upper of two gaps corresponds to a doping of 0.16. These solutions are found within some range of q, but the dips in the DOS are relatively weak. A satisfactory solution is found for q_x and q_y near 0.14 and 0.11, respectively. The average of the two vectors, 0.125, is comparable to the wavelength in the LMTO calculation (where $V_q^m = 8$ mRyd). Two gaps remain in the DOS for larger separation of the two q-vectors, but the dips are found at too high and too low doping. The existence of this multitude of solutions indicates a spread in energy and momentum.

Less doping makes the waves longer, with a narrower waist of the spectrum. The narrowing should be noticed at all energies, even in the absence of SPC at the highest E. Smaller q-vectors have been observed in LBCO for $x = 1/8$ [12] and recently in lightly doped $La_{1.96}Sr_{0.04}CuO_4$ [24], in agreement with this prediction. However, in the latter case the spin modulation has turned from parallel (to the Cu–O bond) to the diagonal direction. Calculations for waves with this orientation cannot consider waves of realistic length, but the results for short waves show that V_q for movements of oxygens are larger than for movements of Cu or La. This is consistent with the observation of the largest q-vectors for low frequency modes as in [24]. A tendency for shorter q-vectors at larger T can be expected from the T-dependences of u and m, in qualitative agreement with the observations for slightly doped LSCO [24].

The upper part of the spectrum should be insensitive to isotope shifts, since no phonons can enforce spin waves at very high energies. Heavier O-isotopes will decrease the frequencies for the phonons and the coupled spin waves, and move the waist to lower E. A heavier mass M will also decrease u ($u \sim M^{-1/4}$ at low T), which in the model for SPC will make V_q^t smaller. The first expectation is therefore a wider waist at lower energy if heavy O-isotopes are inserted in the planes. However, there are complications since SPC also predicts phonon softening; a phonon coupled to a spin wave will have ~10–20% lower energy than if the spin wave is absent [18]. Thus, if u decreases there will be less softening. In addition, from the correlation between V_q^m and wavelength it is expected that q becomes larger, i.e. moving away from the ideal relation between q and doping if u changes. These two effects will reduce the isotope shifts on the excitation energy. No significant isotope shift on the excitation spectra has been detected so far [25].

In conclusion, these calculations suggest that the hourglass shaped dispersion of the spin wave spectrum is a consequence of different degrees of SPC for different phonon modes. The 'half-breathing' O-mode in the mid-part of the phonon spectrum is found to be the most efficient one, which explains the waist in the spin wave dispersion. The SPC-NFE model simulates several normal state properties surprisingly well [19]. There is nothing special with the electronic band structure in this scenario. The LSDA bands are essentially correct for doped systems, although the exchange enhancement is underestimated in normal LSDA calculations [26].

I am grateful to B Barbiellini and C Berthod for various discussions.

References

[1] Tranquada J M, Sternlieb B J, Axe J D, Nakamura Y and Uchida S 1995 *Nature* **375** 561
[2] Sahrakorpi S, Lindroos M, Markiewicz R S and Bansil A 2005 *Phys. Rev. Lett.* **95** 157601
[3] Damascelli A, Shen Z-X and Hussain Z 2003 *Rev. Mod. Phys.* **75** 473 and references therein
[4] Norman M R, Ding H, Randeria M, Campuzano J C, Yokoya T, Takeuchi T, Takahashi T, Mochiku T, Kadowaki K, Guptasarma P and Hinks D G 1998 *Nature* **392** 157
[5] Uchiyama H, Baron A Q R, Tsutsui S, Tanaka Y, Hu W-Z, Yamamoto A, Tajima S and Endoh Y 2004 *Phys. Rev. Lett.* **92** 197005
[6] Pintschovius L and Braden M 1999 *Phys. Rev.* B **60** R15039
[7] Fukuda T, Mizuki J, Ikeuchi K, Yamada K, Baron A Q R and Tsutsui S 2005 *Phys. Rev.* B **71** 060501(R)
[8] Rubio Temprano D, Mesot J, Janssen S, Conder K, Furrer A, Sokolov A, Trounov V, Kazakov S M, Karpinski J and Muller K A 2001 *Eur. Phys. J.* B **19** 5
[9] Gweon G-H, Sasagawa T, Zhou S Y, Graf J, Takagi H, Lee D-H and Lanzara A 2004 *Nature* **430** 187
[10] Yamada K, Lee C H, Kurahashi K, Wada J, Wakimoto S, Ueki S, Kimura H, Endoh Y, Hosoya S, Shirane G, Birgenau R J, Greven M, Kastner M A and Kim Y J 1998 *Phys. Rev.* B **57** 6165
[11] Hayden S M, Mook H A, Dai P, Perring T G and Doğan F 2004 *Nature* **429** 531
[12] Tranquada J M, Woo H, Perring T G, Goka H, Gu G D, Xu G, Fujita M and Yamada K 2004 *Nature* **429** 534

[13] Vignolle B, Hayden S M, McMorrow D F, Rönnow H M, Lake B and Perring T G 2007 *Nat. Phys.* **3** 163
[14] Piekarz P and Egami T 2005 *Phys. Rev.* B **72** 054530
[15] Eremin I, Kamaev O and Eremin M V 2004 *Phys. Rev.* B **69** 094517
[16] Jarlborg T 2001 *Phys. Rev.* B **64** 060507(R)
[17] Jarlborg T 2003 *Phys. Rev.* B **68** 172501
[18] Jarlborg T 2007 *Physica* C **454** 5
[19] Jarlborg T 2007 *Phys. Rev.* B **76** 140504(R)
[20] Thomsen C and Cardona M 1989 *Physical Properties of High-Temperature Superconductors* ed D M Ginsberg (Singapore: World Scientific) p 409
[21] Humlicek J, Litvinchuk A P, Kress W, Lederle B, Thomsen C, Cardona M, Habenmeier H U, Trofimov I E and König W 1993 *Physica* C **206** 345
[22] Saini N L, Lanzara A, Oyanagi H, Yamaguchi H, Oka K, Ito T and Bianconi A 1997 *Phys. Rev.* **55** 12759
[23] Chen H and Callaway J 1992 *Phys. Rev.* B **46** 14321
[24] Matsuda M, Fujita M, Wakimoto S, Fernandez-Baca J A, Tranquada J M and Yamada K 2008 arXiv:0801.2254v1 [cond-mat]
[25] Pailhes S, Bourges P, Sidis Y, Bernhard C, Keimer B, Lin C T and Tallon J L 2005 *Phys. Rev.* B **71** 220507
[26] Jarlborg T 2004 *J. Phys.: Condens. Matter* **16** L173