Spin-phonon coupling, $q$-dependence of spin excitations and high-\(T_C\) superconductivity from band models.

T. Jarlborg

DPMC, University of Geneva, 24 Quai Ernest-Ansermet, CH-1211 Geneva 4, Switzerland

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 long "1-dimensional" unit cells of La$_2$CuO$_4$ show that the coupling between antiferromagnetic spin waves and phonons is larger for distortions of oxygens than for Cu or La. When this result is applied to a 2-dimensional, free-electron-like band, it leads to a $q$-dependent spin excitation spectrum in good agreement with recent experiments. It is argued that important parameters for spin-phonon coupling, which comes out from the comparison between experiment and theory, are relevant for the mechanism of superconductivity, and are large enough to explain a high $T_C$.

PACS numbers: 74.25.Jb,74.20.-z,74.20.Mn,74.72.-h

Many measurements have revealed the existence of complex superstructures and pseudogaps in high-$T_C$ copper oxides. A picture of spatial, stripe-like regions with excitations of charge and spin modulations emerged quite early from neutron scattering experiments [1]. Angular resolved photoemission confirms that the bandstructure agree well with calculations, where one dispersive band gives rise to an ubiquitous barrel-like Fermi-surface (FS) (or a 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 [5,6,7], 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 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 "hour-glass" shaped (\(q,\omega\))-dispersion, even at non-superconducting compositions [11,12,13]. Here, it is shown that the characteristic spin dispersion can be understood in terms of spin-phonon coupling (SPC), and secondly it is concluded that phonons promote equal spin pairing and a large \(\lambda_{sf}\) for spin fluctuations, and therefore SPC is important for the mechanism of superconductivity.

Previous ab-initio band calculations for supercells containing 1-dimensional (1D) phonon and spin-wave modulations in the CuO bond direction ([1,0,0]), show large SPC within the CuO plane of these systems [14]. This means that an antiferromagnetic (AFM) wave of the correct wave length and the proper phase is stronger when it co-exists with the phonon [15]. These results, in combination with 2D free-electron like bands, have been used for modeling of many normal state properties of the high-$T_C$ materials [16,17]. Phonon softening, dynamical stripes, correlation between \(q\) 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. The present calculations consider SPC between spin waves and four different types of phonon distortions.

First, we discuss the ab-initio band calculations made for La$_{2-x}$Ba$_x$CuO$_4$ (LBCO), where the virtual crystal approximation (VCA) is applied to La-sites to account for the doping. The calculations are made for long supercells oriented along the CuO bond direction, by using the linear Muffin-Tin Orbital method (LMTO) in the local spin-density approximation (LSDA), as has been described previously [15]. 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 \(K_u = d^2E/dp^2\), \(E\) is the total energy, and \(p = u \text{ or } m\), respectively [15]. The force constants \(K_u\) for the different atoms are taken from measurements on YBa$_2$Cu$_3$O$_7$ [17,18]. The resulting \(u/\omega_0\), where \(\omega_0\) is the lattice constant, are shown in the Table for \(T \approx 100\,K\) (the interesting temperature range for \(T_c\) and \(T^*\)). An approximate calculation of \(K_m\) for a short wave in HgBa$_2$CuO$_4$ corresponds to magnetic moments \(m\) on Cu of about \(\sim 0.09\mu_B\) at 100 K [19]. Such moments are typically obtained by application of a magnetic field of the order of \(\pm 5\) mT [15].

Results of calculations are presented here showing a varying degree of SCP for different phonons. These calculations consider distortions in a cell with 16 formula units (8\(a_0\) along \(\bar{x}\)) with displacements of La and apical O along \(\bar{z}\), and Cu and planar-O along \(\bar{x}\), and the phonons may co-exist with AFM spin-waves. 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 are moved 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. The calculated results for the maximal potential shifts on Cu caused by phonons (∇\(V_q\)) and by spin waves (∇\(V^m_q\)) in the spinpolarized potential) for the 4 types of movements in the cell are shown in the Table, together with the result for \(V^m_q\) with-
out phonon. That the phase between the phonon and spin wave is crucial for having a positive SPC is shown by the fact that \( V_{q}^{p} \) for an "apical-oxygen" phonon is reduced from 10 to below 8 mRy when the phase between the phonon and the spin wave is shifted by \( \pi \).

Measured \( V_{q}^{m} \) and \( V_{q}^{p} \) and calculated \( V_{q}^{m} \) and \( V_{q}^{p} \) phonon frequencies for zone boundary phonons for \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) are indicative for typical \( \hbar \omega \) for each atomic character of the phonon DOS. Phonon energies and partial phonon DOS for each site, \( N_{\text{site}} \), have been calculated by Chen and Callaway \( \text{[23]} \) for a similar cuprate system, \( \text{Nd}_2\text{CuO}_4 \), and the same \( N_{\text{site}} \) can be assumed to be representative for LBCO. From these references it is seen that 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 \( \text{[17, 21, 23]} \). Our estimations of \( N_{\text{site}} \) from ref. \( \text{[23]} \) are shown in the last part of Table I. A rescaling is made for the wavelength. The LMTO results are calculated for short waves, \( 8a_0 \), but the waves are longer at the doping \( x = 0.16 \) (corresponding to \( q \approx 0.08 \)), when \( V_{q}^{p} \) and \( V_{q}^{m} \) are larger by factors of \( \sim 1.1 \) and \( \sim 1.5 \), respectively \( \text{[16]} \). These factors might be fine tuned later if the calculated wave lengths fall outside the expected range, but in the first calculations we calculate the total \( V_{q}^{t} \) from,

\[
V_{q}^{t} = \sum_{i} (1.1 \ast V_{q,i}^{p} + 1.5 \ast V_{q,i}^{m}) \ast N_{i}
\]

(1)

where \( i \) is the site index. The resulting \( V_{q}^{t} \) are 17, 18, 23 and 22 mRy at the energies centered around 15 (La), 25 (Cu), 50 (plane-O) and 60 meV (apical-O), respectively. These values are used in the subsequent calculations of the \( q - \omega \) dispersion. As discussed later, spin waves at higher energy are independent of the 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, \( V(\vec{x}) = V_{q}^{t} \exp(-i\vec{Q} \cdot \vec{x}) \) (and equivalently with \( \vec{y} \) 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 \( \vec{Q}/2 \). A further modulation \( (\vec{q}) \) of this order into 1D-stripes perpendicular to \( \vec{x} \) (or "checkerboards" in 2D along \( \vec{x} \) and \( \vec{y} \)) is achieved by a multiplication of the potential by \( \exp(i\vec{q} \cdot \vec{x}) \), where \( \vec{q} < \vec{Q} \). Totally, this makes \( V(\vec{x}) = V_{q}^{t} \exp(-i\vec{Q} \cdot \vec{x}) \), where \( Q_k = Q - q \). The periodicity of this potential is now larger, and the gap moves away from \( \vec{Q}/2 \) to \((\vec{Q} - \vec{q})/2\). Magnetic side spots appear at \( \vec{q}/2 \) surrounding \( \vec{Q}/2 \) in probes which can separate the two periodicities. A 3x3 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 \( V_{q}^{0} = 0 \)) contains 2 electrons up to \( E_F \sim 0.15 \text{Ry} \), when the effective mass is one. The total band width is quite close to the real band. The model is entirely 2-D, (the DOS is constructed from a sum over all states in the plane), which is reasonable in view of the very small band dispersion along \( k_x \) of real band structures for the cuprates \( \text{[14]} \).

An important result of the 2D-NFE model is that it leads to a correlation between doping and the amplitude of \( V_{q}^{t} \) \( \text{[16]} \). 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 vary the q-vector until it fits. The present calculations are made for 0.16 holes per Cu (which is close to \( x \) in ref. \( \text{[13]} \)) with \( V_{q}^{t} \) from eq. 1.

The results are shown in fig. 1, with positive and negative \( q \) displayed symmetrically, and the spectrum is shaped like an hour-glass with a "waist" at intermediate energy. The points below 70 meV are for the coupling to the 4 types of phonons. The general shape of the \( q - \omega \) dependence and the amplitudes of \( q \) are similar as in ref. \( \text{[13]} \), 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. The calculated results depend on the different parameters in the NFE model. Larger band mass leads to smaller amplitudes of \( q \). The variations of \( q \) is a result of the differences in \( V_{q}^{t} \) at different energies. Consequently, if no mixing of the phonon modes (through the \( N_{\text{site}-\text{coefficients}} \)) were made it would increase the variations of \( q \), even though the general hour-glass shape would remain.

The uppermost part of the spectrum needs some comments. Spin waves and phonons are tied together at the same frequency and q-vector for the optimal mechanism of SPC. Spin waves with higher frequency than the phonons cannot profit from this mechanism. Thus, it is assumed that spin waves with frequencies higher than the phonon frequencies are independent of lattice vibrations, whereby \( V_{q}^{p} = 0 \). This "phonon independent" result is put rather arbitrarily at \( \hbar \omega \sim 120 \text{meV} \) in fig. 1, which

| wave | no-phon pl-O_2 | La_s | ap-O_2 | Cu_s |
|------|----------------|------|--------|------|
| \( u/a_0 \) | - | 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 |
| \( N_{\text{La}} \) | - | 0.0 | 0.65 | 0.0 | 0.35 |
| \( N_{\text{ap-O}} \) | - | 0.35 | 0.0 | 0.65 | 0.0 |
| \( N_{\text{Cu}} \) | - | 0.2 | 0.2 | 0.0 | 0.6 |
is about twice the highest phonon frequency. The calculated wavelengths for the SPC modes below ~70 meV are found to be longer than the length of the supercell in the LMTO calculation, which justifies the factors 1.1 and 1.5 in eq. 1. However, the solution without SPC leads to a wave which is even shorter than the cell in the LMTO calculation. The the factor 1.5, in eq. 1, is not justified for this (phonon free) mode, and a corrected calculation with \( V_q^t = 8 \) mRy is more appropriate, with a solution for \( q = 0.24 \), see fig. 1. In contrast to the cases with SPC and larger \( V_q^t \), this solution implies a very short wave, 4-5 \( a_0 \) only, where \( V_q^t \) might be reduced even more. This will, because of the self-consistent feed-back between spin density and potential, lead towards a vanishing spin wave [15]. 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 \( y \) [16], 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 \( V_q^m = 8 \) mRy). 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 that large broadening and damping is expected at high energies. Such fluctuations are spread in energy and momentum. They decay rapidly, since they are not linked to the phonon spectrum.

Less doping will make the waves longer and the waist in the \( q-h\omega \) diagram becomes narrower. The narrowing should be noticed at all energies even at the highest \( E \) where spinwaves are excited without the help of phonons. Smaller \( q \)-vectors have been observed recently in lightly doped La_{1−\delta}Sr_{\delta}CuO_{4} [24], in agreement with this prediction. However, the spin modulation has turned from parallel (the Cu-O bond) to diagonal direction at such low doping. In addition, the waist in the observed spectrum is found at lower energy than for parallel doping [24], which in the SPC-model implies increased SPC for phonons at lower energy. Ab-initio calculations for modulations along the diagonal direction are needed to verify if La- or Cu-modes grow in importance relative to the high energy O-modes.

Heavier O-isotopes will decrease the frequencies for the phonons and the coupled spin waves, and move the waist to lower \( E \). They may also decrease \( u \), at least at low temperature, which in the model for SPC will make \( V_q^t \) smaller. From the correlation between \( V_q^m \) and \( q \) wave length it is expected that \( q \) becomes larger. In all, this leads to a wider waist at lower energy, while the upper part of the spectrum should be insensitive to isotope shifts.

Undoped cuprates are generally stable AFM insulators. The exchange enhancement for spin waves in doped cuprates is moderately large in the absence of phonons, and it becomes peaked if a phonon can intervene. This can be translated into a large phonon deformation potential. The unusual part of it is that this deformation potential resides in the equal spin part, and hence it is more correct to associate this deformation potential with a large \( \lambda_{sf} \) for spin fluctuations. The calculated \( V_q \)-parameters determine the spin excitation spectrum for large SPC, but they are also the key parameters for estimates of superconducting \( T_c \). For instance, \( V_q^p \) corresponds to the monopolar matrix element for electron-phonon coupling, \( \lambda_{ph} \). The good agreement between the calculated spin excitations and experiment suggests that the calculated \( V_q^t \)'s are of the correct order. It is not clear how the contribution to \( T_c \) from coexisting phonon and spin fluctuations will be shared, but a simple account for separate \( \lambda_{ph} \) and \( \lambda_{sf} \) gives rather large values for \( T_c \). The estimates are based on \( \lambda = N \eta^2 / K_u \), where \( N \) is the DOS at \( E_F \) (\( \approx 0.5 (eV \cdot Cu\cdot spin)^{-1} \)), and \( I = (\Delta V^p/\Delta u) \). From the Table and with \( a_0 = 3.78A, \Delta V^p/\Delta u \sim 3.8eV/A \) for a pure (plane) O-phonon, which makes \( \lambda_{ph} \approx 0.5 \), for \( K_u \) of the order 15 eV/A^2. If phonons stir up a spin fluctuation (through SPC) one should only consider the equal spin part in \( \Delta V^m \), which is almost of the same order as \( \Delta V^p \) for the O-phonon (see Table). Moreover, one should add a \( K_m \) to \( K_u \) because of the energy of the spin wave, but this contribution can be negative if the coexistence makes the phonon softer (\( K_m \) without phonon may be large and positive). By assuming \( K_m \sim 0 \) this gives \( \lambda_{sf} \approx 0.4 \), and it will increase faster than \( \lambda_{ph} \) when the doping becomes smaller. The BCS formula \( T_c = 1.13 \cdot h \omega \cdot exp (-1/\lambda) \) makes \( T_c \) of the order 80-40 K, for these two \( \lambda \)'s, when \( h \omega = 50meV \).
This is very approximate, but it shows that the coupling strength can be sufficient for a large $T_c$. Note also that since the exchange enhancement in LSDA is too weak to stabilize AFM in undoped LSCO, it will also produce a too low value of $\lambda_{sf}$.

The total SPC becomes too large at underdoping when the wave modulations are longest, so $\lambda$ and $\lambda_{sf}$ are reduced because of the pseudogap, and hence the pseudogap competes with superconductivity. On the overdoped side $\lambda$ is reduced because of a small $\Delta V$, and $T_c \rightarrow 0$. Optimal conditions for a high $T_c$ are found at intermediate doping, but the estimates are not precise enough for finding the exact value or the exact doping limits for vanishing $T_c$. The role of phonons decreases as $x \rightarrow 0$, since the ratio $V^m/V^p$ is growing at small doping.

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 midpart 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. There is nothing special with the electronic band structure in this scenario. The LDA bands are essentially correct for doped systems, although the exchange enhancement is underestimated in normal LSDA calculations. Finally, it is argued that the observed spin excitations provide important information about the mechanism of superconductivity, since the excitation spectrum can be related directly to the coupling parameters. In particular, there is an unusual contribution to the equal spin pairing parameter $\lambda_{sf}$ coming from the coupling to phonons.

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

[1] J.M. Tranquada, B.J. Sternlieb, J.D. Axe, Y. Nakamura and S. Uchida, Nature 375, 561 (1995).
[2] S. Saharikorpi, M. Lindroos, R.S. Markiewicz and A. Bansil, Phys. Rev. Lett. 95, 157601 (2005).
[3] A. Damascelli, Z.-X. Shen and Z. Hussain, Rev. Mod. Phys. 75, 473, (2003) and references therein.
[4] M.R. Norman, H. Ding, M. Randeria, J.C. Campuzano, T. Yokoya, T. Takeuchi, T. Takahashi, T. Mochiku, K. Kadowaki, P. Guptasarma and D.G. Hinks, Nature 392, 157, (1998).
[5] H. Uchiyama, A.Q.R. Baron, T. Tsutsui, Y. Tanaka, W.-Z. Hu, A. Yamamoto, S. Tajima and Y. Endoh, Phys. Rev. Lett. 92, 197005, (2004).
[6] L. Flintschovius and M. Braden, Phys. Rev. B60, R15039, (1999).
[7] T. Fukuda, J. Mizuki, K. Ikeuchi, K. Yamada, A.Q.R. Baron and T. Tsutsui, Phys. Rev. B71, 060501(R), (2005).
[8] D. Rubio Temprano, J. Mesot, S. Janssen, K. Conder, A. Furrer, A. Sokolov, V. Trounou, S.M. Kazakov, J. Karpinski, and K.A. Muller, Eur. Phys. J. B19, 5 (2001).
[9] G.-H. Gweon, T. Sasagawa, S.Y. Zhou, J. Graf, H. Takagi, D.-H. Lee and A. Lanzara, Nature 430, 187, (2004).
[10] K. Yamada, C.H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shigane, R. J. Birgenau, M. Greven, M.A. Kastner and Y.J. Kim, Phys. Rev. B57, 6165, (1998).
[11] S.M. Hayden, H.A. Mook, P. Dai, T.G. Perring and F. Doğan, Nature 429, 531 (2004).
[12] J.M. Tranquada, H. Woo, T.G. Perring, H. Goka, G.D. Gu, G. Xu, M. Fujita and K. Yamada, Nature 429, 534 (2004).
[13] B. Vignolle, S.M. Hayden, D.F. McMorrow, H.M. Rönnow, B. Lake and T.G. Perring, Nature Physics 3, 163, (2007).
[14] T. Jarlborg, Phys. Rev. B64, 060507(R), (2001).
[15] T. Jarlborg, Physica C454, 5, (2007).
[16] T. Jarlborg, Phys. Rev. B76, 140504(R), (2007).
[17] C. Thomsen and M. Cardona, in "Physical Properties of High-Temperature Superconductors": ed. D.M. Ginsberg (World Scientific, Singapore, 1989) 409.
[18] J. Humlicek, A.F. Litvinchuk, W. Kress, B. Lederle, C. Thomsen, M. Cardona, H.U. Habenmeier, I.E. Trofinov and W. König, Physica C 206, 345 (1993).
[19] T. Jarlborg, Phys. Rev. B68, 172501 (2003).
[20] T. Jarlborg, Phys. Lett A295, 154 (2002).
[21] R.E. Cohen, W.E. Pickett and H. Krakauer, Phys. Rev. Lett. 64, 2575, (1990).
[22] O.K. Andersen, A.I. Liechtenstein, O. Rodriguez, I.I. Mazin, O. Jepsen, V.P. Antropov, O. Gunnarsson and S. Gopalan, Physica C185-189, 147, (1991).
[23] H. Chen and J. Callaway, Phys. Rev. B46, 14321, (1992).
[24] M. Matsuda, M. Fujita, S. Wakimoto, J.A. Fernandez-Baca, J.M. Tranquada and K. Yamada, cond-matt arXiv:0801:2254v1 (2008).
[25] T. Jarlborg, J. Phys.: Cond. Matter, 16, L173, (2004).