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Electron mechanism for the tilting transition in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

S. Barisić and I. Batistić*

Department of Physics, Faculty of Science, P.O.B. 162, Marulicev trg 19, 41001 Zagreb, Croatia, Yugoslavia

*Institute of Physics of the University, P.O.B. 304, Bijenička c.46, 41001 Zagreb, Croatia, Yugoslavia

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Résumé. — Nous montrons que la déformation qui fait onduler les plans CuO₂ est couplée aux électrons par l'intermédiaire de la variation des éléments de matrice Umklapp dans l'interaction électron-électron. Ces derniers ne sont importants que pour de faibles concentrations de Sr, si les interactions, supposées alors répulsives, sont suffisamment petites. Le premier terme dans la contribution à l'énergie de déformation est quadratique en déformation et diminue de façon critique la fréquence des modes d'ondulation lorsque la température décroît. La résistivité, bien qu'augmentée par Umklapp, est continue à travers la transition à la phase ondulée. Notre modèle ressemble quelque peu à celui développé précédemment pour les supraconducteurs organiques. Nous discutons aussi brièvement les propriétés du modèle qui concernent la supraconductivité à $T_c$ élevée.

Abstract. — It is shown that the tilting deformation is coupled to the electrons through the tilting induced variation of the Umklapp matrix elements. The latter are important only for small concentrations of Sr if the electron-electron interaction, (which are assumed to be repulsive), are reasonably small. The leading term in the resulting deformation energy is quadratic in the tilting deformation and leads to the critical decrease of the tilting mode frequency when the temperature is lowered. The resistivity, although enhanced by Umklapp, is continuous through the tilting transition. The model bears some resemblance to that developed previously for $4k_F$ deformations in organic superconductors. Some consequences of the model for the high $T_c$ superconductivity are briefly discussed.

Recent measurements [1,2] of the lattice properties have shown that the orthorhombic to tetragonal transition, at $T = 423$ K in $\text{La}_2\text{CuO}_4$, is driven by the $(0.5, 0.5,0)$ tilting mode $\eta$, depicted [3] in figure 1. Actually it turned out [1,2] that $\eta^2 \sim (T - T)^{2\beta} \sim \epsilon$ where $\epsilon$ is the $(0,0,0)$ shear of the basal CuO₂ plane. $\epsilon$ appears therefore only as a secondary order parameter induced by the tilting $\eta$, through the $\eta^2$ Landau term. This conclusion is corroborated by the study [2] of the pretransitional softening of the phonon modes in monocrystals of $\text{La}_2\text{CuO}_4$. The described result is important provided that the instability of the tilting mode reflects the properties of the electron system. The understanding of the tilting transition as the one which occurs at the highest temperature may then shed some light on the nature of other transitions, [4,5] including superconductivity [6].

There are some indications that the electrons are the source of the tilting instability. First, $\tilde{T}$ is of the same order of magnitude as the transition temperature of electronic transitions to the antiferromagnetic [4,5] (AF) and not so far from the superconducting [6] $T_c$. Second the phonon softening is rather well localized [1] in
the Brillouin zone, suggesting large correlation lengths. Third, $T$ is strongly dependent [1,7,8] on the doping $x$ in e.g. $La_{2-x}Sr_xCuO_4$. The tilting disappears at $x \approx 0.2$. On the other hand it was recognized [9,10] very early that $\eta$ is not coupled linearly to the band electrons, and therefore any kind of straightforward Peierls transition is ruled out. This fact even led to the suggestions that the tilting is not electronically driven, disregarding its strong dependence on doping.

\begin{align}
\Delta_{pd} &= \frac{\Delta_{pd}}{4t} \\
\delta &= \frac{\Delta_{pd}}{4t} \approx 0.2.
\end{align}

where $\delta = \Delta_{pd}/4t$. It is usually believed [12] that $\delta$ is small, $\delta<1$. The corresponding band structure is exhibited in figure 3. This figure shows that the Fermi surface can be divided in the van Hove (vH) singularities, where the electron velocity vanishes, and the quasi one-dimensional (1d) sections in between. The electron which corresponds to those latter states are quite mobile and propagate along the diagonals of the CuO$_2$ plane.

Let us next turn to interactions. Bearing in mind that the pure system exhibits AF ordering [4,5] we choose repulsive Coulomb interaction. The latter are conveniently parametrized in terms of the Hubbard matrix elements $U_d$ and $U_p$ for the Cu and O sites respectively and the intersite matrix element $V$, while the long range Coulomb forces can be taken as screened out [13]. In the band representation of figures 2 and 3 the interaction matrix elements are then given as linear combinations of the $U_d$, $U_p$ and $V$. However, not all of these combinations are important. In the weak coupling limit $(V, U_p, dW)$ only the matrix elements which belong to the Hamiltonian $H_B$ of the upper sub-band may play an essential role. Among the intraband Umklapp matrix elements we need only those involved in electron-hole diagrams, which appear in the parquet or ladder approximations. Taking $k$'s of $U(k_1, k_2, k_3, k_4)$, with $k_1+k_2=k_3+k_4+G$ in the notations of figure 3 to the representative terms are (ignoring $V$ for simplicity)

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Tilted CuO$_6$ octahedron within the cage of La. The arrows indicate the shifts of the oxygen atoms.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{Densities of states of the lower and upper sub-bands for $\delta = \Delta_{pd}/4t \approx 0.2$. The gap between sub-bands is equal to $\Delta_{pd} = E_d - E_p$.}
\end{figure}
\[ U_U(A B C D) = U_d \cos^4 \varphi \] (2a)

\[ U_U(a b c d) = U_d \cos^4 \varphi - 2U_p \sin^4 \varphi \] (2b)

where

\[ \tan \varphi = \sqrt{1 + \delta^2 - \delta} \] (3)

with \( \delta = \Delta_{pd}/4t \). The Umklapp (2a) relating the vH electrons depends only on \( U_d \), since \( U_p \) is removed by interference effects. In contrast to that the Umklapp (2b) which couples the quasi 1D electrons depends on both \( U_d \) and \( U_p \). Unlike the Umklapp (2a) the Umklapp (2b) may even vanish for special of \( U_d \) and \( U_p \) and in particular for \( U_d = 2U_p \) at \( \delta = 0 \) when the gap \( \Delta_{\text{gap}} \) between the sub-bands in figure 2 is closed. Postponing the description of the physical meaning of this cancellation let us turn now to the evaluation of the dependence of \( U_U \) on \( \eta \). To this end we note that both Umklapps (2) are linear in \( \delta \) for \( \delta < 1 \).

![Fig. 3.— Brillouin zone of the upper sub-band. The capitals A, B, C and D indicate the regions of the vH singularities, while letters a, b, c, and d correspond to the flat 1D-parts of the Fermi surface.](image)

Apparently both Umklapps (2) depend on \( \eta \) either through \( \Delta_{pd} \) or through \( t \). The symmetry requires this dependence to be quadratic in both cases. Indeed, considering \( \Delta_{pd} \), the tilt induced rotation of the CuO\(_4\) octahedron (Fig. 1) in the crystal field of the La\(^{3+}\) cage decreases the site energy \( E_b \) of the in plane O's which move towards La\(^{3+}\). The effect can be evaluated using the generalized Evjen-Frank method, [14]

\[ \tilde{\Delta}_{pd} = \Delta_{pd} + \frac{3\sqrt{2} \epsilon^2}{4} \frac{1}{a} \eta^2, \] (4)

where for simplicity we have disregarded the difference between the lattice constants \( c \) and \( a \). The corresponding variation of \( t \) can be found by expanding \( t \) in the conventional way. Inserting those variations in equation (3) and then in equation (2) the Umklapp matrix elements (2a,b) can easily be found at \( \delta < 1 \) as

\[ \tilde{U}_U^{(i)} = U_U^{(i)} + \tilde{U}_U^{(i)} \eta^2 \] (5)

with \( i = A, a \) respectively. \( \tilde{U}_U \) is in general large due to the large prefactor of \( \eta^2 \) in equation (4). In addition \( \tilde{U}_U^{(A)} \) is always positive. Even \( \tilde{U}_U^{(a)} \) is of the right sign in the large part of the parameter space \( (U_d, U_p, \delta) \). \( \tilde{U}_U > 0 \) is essential for the understanding of the tilting transition, as will be argued now.

The harmonic deformation energy which corresponds to equation (5) can be conveniently determined under two assumptions. First, the Born-Oppenheimer adiabatic approximation will be assumed to hold when \( \eta \) is varied. Second, \( \tilde{U}_U \) will be considered as a relevant parameter, i.e. that it controls the singular behaviour of different correlators which define the average value \( \langle \tilde{H}_B \rangle \) of the electron intra-band Hamiltonian \( \tilde{H}_B \). The development of those singularities decreases \( \langle \tilde{H}_B \rangle \) and the increase of \( \tilde{U}_U \)

can then only decrease \( \langle \tilde{H}_B \rangle \). In other words \( \partial \langle \tilde{H}_B \rangle / \partial \tilde{U}_U < 0 \). The \( \eta \) induced variation of the electron energy \( \langle \tilde{H}_B \rangle \) is thus negative provided that \( \tilde{U}_U > 0 \). As a result the frequency \( \tilde{\omega} \) of the tilting mode is decreased according to

\[ \tilde{\omega}^2 = \tilde{\omega}_0^2 - \frac{\tilde{U}_U}{m a^2} \left| \frac{\partial \langle \tilde{H}_B \rangle}{\partial \tilde{U}_U} \right|, \] (6)

where \( \tilde{\omega}_0 \) is its bare value.

The Umklapp term is relevant only when the correlator [15] which multiples it in \( \langle \tilde{H}_B \rangle \) is itself singular. When this singularity is dominant its contribution to \( \langle \tilde{H}_B \rangle \) can be evaluated...
in two steps [16]. In the first step the Umklapp part of $\hat{H}_B$ is treated in second order perturbation theory. The resulting convolution $\tilde{\chi}_4^{(0)}$ of the two singular electron-hole bubbles is next renormalized [15-17] to $\tilde{\chi}_4$ by inserting the leading logarithmic corrections, which involve all interactions and the Umklapps (2) in particular. $\tilde{\chi}_4$ describes the pd valence fluctuations in the limit $\delta < 1$ when the Umklapps (2) are linear in $\delta$. This can be understood on noting that $\Delta_{pd}$ enters the original Hamiltonian as the external staggering field which is linearly coupled to the site density $n_p$. In the corresponding band picture the external $\delta$ is linearly coupled to $\tilde{\chi}_4$ when the Umklapps (2) are linear in $\delta$ (\(\delta < 1\)) i.e. $\tilde{\chi}_4$ is the susceptibility to $\delta$. In addition, the values of the Umklapps (2) at $\delta = 0$ add to this $\delta$ an internal, interaction-induced staggering field. As discussed in connection with equations (2) and (3) this field is in general finite, i.e. it can vanish only in equations (2b) at $U_d = 2U_p$.

The softening of the tilting frequency (6) reflects then directly the increased susceptibility $\tilde{\chi}_4$ of the system to the (external and internal) staggering field, i.e. the pd valence fluctuations are enhanced. As the effective charge of the 0 site is increased the 0 ions are attracted towards the positive La$^{3+}$ sites and the tilting occurs.

The experiments show that the dependence of $\omega(q)$ on $[1,2] T, q$ and $[1,2,7,8] x$ is appreciable but not dramatic, which is consistent with the weak coupling approach used here. Actually, whatever perturbation theory (parquet or ladder) is used in the evaluation of $\partial (H_B)/\partial U_U$ the associated high temperature length scale should be given by elementary diagrams, namely $\xi = aW/T$. Smaller scales can occur only below some electronic phase transition or crossover.

The bare $\xi$ describes reasonably well the width of the observed anomaly $\Delta k \approx 10^{-1} a^{-1}$, and the concentration $x_e$ which suppresses the tilting, $x_e \approx a \xi^{-1} \approx 10^{-1}$. Indeed the doping $x$ introduces the holes in the upper subband of figure 2. The resulting shift of the Fermi energy separates the normal and Umklapp contributions of the electron-hole diagrams. In addition, both log and $(\log)^{2}$ contributions of respectively, the quasi 1d parts and the van Hove singularities are depleted. As a result the Umklapps cease to be relevant. The effect occurs on the scale $\Delta k_F \approx \xi^{-1}$.

Note that the electron redistribution frequency can by found in the same way as $\xi$. It is equal to $T$. Since $\tilde{T} > \tilde{\omega}_0$ this justifies the use of the adiabatic approximation in deriving equation (6).

It should be further pointed out that the electronic properties are predicted to be continuous through the tilting transition. This is conveniently illustrated with the help of $\tilde{\chi}_4$, but holds also in the more general case of equation (6). According to equation (6) $\tilde{\chi}_4$ is finite at $\tilde{T}$. Below $\tilde{T}$ only the average values of umklapps (2) are affected by the development of $\eta$. To the second order this effect is given by equation (5). The variation of Umklapps is in principle reflected in all correlation functions including $\tilde{\chi}_4$. However the effect is smoothed out when $U_U$ is finite in equation (5). In particular only a weak crossover effect is expected to occur at $\tilde{T}$ in the resistivity, in good agreement with observations [2]. $\tilde{\chi}_4$ should continue to increase below $\tilde{T}$ heading towards the Mott metal to insulator transition in $\tilde{H}_B$. The actual evaluation of $\tilde{\chi}_4$ or of the other correlators in $\left\langle \tilde{H}_B\right\rangle$ requires at least the solution of the parquet problem. The latter was discussed [18,19] for layered oxides, retaining only the contributions of the van Hove singularities. In particular this amounts to disregarding $\chi_4^{(A)}$ in $\chi_4$. $\chi_4^{(A)}$ was not explicitly considered although the low temperature enhancement of the renormalized Umklapp (2a) indicates that $\chi_4^{(A)}$ diverges at low $T$. Unfortunately it was assumed [18,19] that $\Delta_{pd} \gg t$ (i.e. $\cos^4 \phi \approx 1$ in Eq. (2a)), the limit in which $\eta$ is only weakly coupled to the electrons ($\tilde{U}_U^{(A)} \neq 0$ in equation (5) and subsequently). The many-body analysis of the $U_U^{(A)} \neq U_d$ case is therefore desirable.

It is also difficult to include the contribution of the quasi 1d electrons. However if these electrons can be treated separately from those in vH singularities (which is not obvious) the problem is solvable again and, in particular, the correlation function $\chi_4^{(A)}$ can be found [15]. In addition it is instructive to discuss the 1d problem in some detail. Keeping thus only the 1d contribution equation (2b) it can be easily seen that the latter corresponds [20] to the contribution of an alternating chain of d and p sites with site energies $E_d = E_3, E_p = E_p$, Coulomb energies $U_d = U_d$ and $U_p 2U_p$ and the overlap $t = \sqrt{2}t$. For $\Delta_{pd}$ small ($\delta < 1$) and $U_d = U_p = U$ this model maps on that for the Bechgaard organic metals [21]. Indeed in this case equation (2b) reduces to $U_U^{(A)} = \tilde{U} \Delta_{pd} \sqrt{2}t$ characteristic [22] of those
materials. For $\Delta_{pd} \neq 0$ and $U_d \neq U_p$ the model describes the hypothetical mixed stack conductors. It is interesting to note with layered oxides in mind, that even when $[12] \Delta_{pd} = 0 U^{(a)}_U$ of equation (2b) (with $U_p = 2U_p$) is finite provided that $U_d \neq U_p$. Already in the weak coupling limit $U_d, U_p < W$ the chain is dimerized on the level of interactions $(U^{(a)}_U \neq 0)$ even when there is no doubling of the unit cell in the band structure $(\Delta_{pd} = 0)$. The behaviour of the correlation function $\chi_4^{(a)}$ is controlled by $U^{(a)}_U$ whatever the structure of $U^{(a)}_U$ in terms of $U_d, U_p$ and $\Delta_{pd}$. However only for $\delta <1$ does the enhancement of $\chi_4^{(a)}$ lead to a charge distribution on the $p$ and $s$ sites which differs appreciably from the band values.

In spite of this analogy between the layered oxides and organic metals the enhancement of $\chi_4^{(a)}$ in the latter case is accompanied by the temperature dependent dimerization $a_1 - a_2$ of the $pd$ distances rather than by tilting. In the Bechgaard salts $\Delta_{pd}$ is the bond rather than the site gap. The dimerization of distances adds a term linear in $a_1 - a_2$ to this dimerization gap. This contrasts with equation (4) where the dependence on deformation is quadratic. As a result a (small) temperature or composition dependent dimerization is always present in Bechgaard salts [17]. A similar dimerization in layered oxides would change the gap according to $\Delta^2 = \Delta_{pd}^2 + \Delta_a^2$ where $\Delta_a$ is linear in $a_1 - a_2$. As long as the site gap $\Delta_{pd}$ is large with respect to the bond gap $\Delta_a$ this is analogous to equation (4). However the $q=0$ dimerization mode in layered oxides has a bare frequency much larger than $\omega_0$ in equation (6) and it is therefore reasonable that this mode remains stable at $T$.

The enhancement of the renormalized Coulomb interactions in the pure $x = 0 \text{La}_2\text{CuO}_4$ goes presumably into the screening [24,18] in the metallic $x > 0.1$ case, analogous to that introduced in the conventional theory of superconductivity. The interactions mediated by the low frequency phonons [10] or other excitations may then become important. Thus the present description of the tilting transition in terms of the Coulomb interactions does not necessarily imply that the Coulomb interactions alone explain the high $T_c$ superconductivity, although such models have been proposed for Bechgaard salts [21]. It is merely suggested here that the phase diagram observed in layered oxides for $x$ small, analogous in many respects to that of Bechgaard salts, can be reasonably explained in terms of moderate or even weak Coulomb interactions.

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