Краткие сообщения ОИЯИ №4 [30]-88   *JINR Rapid Communications No.4 [30]-88*
УДК 538.945

A THEORETICAL STUDY OF ULTRASONIC ANOMALIES IN $La_{2-x}Sr_xCuO_4$

V.L.Aksenov, S.Flach, N.M.Plakida

A microscopical model for description of acoustic properties at structural phase transition in $La_{2-x}Sr_xCuO_4$ superconductors is proposed. Experimentally observed acoustic anomalies are described in the framework of the model.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

Микроскопическая теория акустических аномалий в $La_{2-x}Sr_xCuO_4$

В.Л.Аксенов, С.Флах, Н.М.Плакида

Предложена микроскопическая модель, описывающая влияние структурного фазового перехода в лантановых оксидных сверхпроводниках на их упругие свойства. Дано объяснение наблюдаемым экспериментально акустическим аномалиям.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Recently a number of investigations of ultrasonic attenuation and elastic constants in $La_{2-x}Sr_xCuO_4$ was done /1-5/. The measured anomalies in the sound velocity are expected to be caused by a structural phase transition (SPT) D_{4h}^{17} - D_{2h}^{18} at $T \neq T_0$ /4/ . This tetragonal-to-orthorhombic SPT results from the softening of the tilting mode (CuO_6 — octahedra rotations around (± 1, 1, 0)-axis) at the X-point of the Brillouin zone (BZ) denoted by wave vectors $\vec{q}_1$ and $\vec{q}_2$ /6/. A model for the microscopic description of this SPT was given in /6/. Thus, the strong $T_0(x)$ — dependence was proposed and a good agreement was reached with results of inelastic neutron scattering experiments /5/.

As was shown in /6/ , the SPT can be described by the motion of oxygen ions using the Hamiltonian:

$$H = \sum_{\ell,k} \frac{m}{2} \dot{X}^2(\ell,k) + \frac{1}{4} \sum_{\ell,k} B X^4(\ell,k) + \frac{1}{2} \sum_{\substack{\ell,\ell' \\ k,k'}} \phi_{kk'}(\ell,\ell') X(\ell,k) X(\ell',k') , \qquad (1)$$

where X(l, k) are the displacements of the k-th oxygen ion in the l-th unit cell along the z-axis (for details we refer to /6/).

Using the transformation

$$X(\ell,k) = \frac{1}{2}\frac{1}{\sqrt{2m}}[\vec{\xi}_k \times (\vec{R}(\vec{\ell}+a\vec{\xi}_k) - \vec{R}(\vec{\ell})]_z \tag{2}$$

with $\vec{\xi}_1 = (1, 0, 0)$, $\vec{\xi}_2 = (0, 1, 0)$ we introduce the octahedron rotation coordinates R_λ ($\lambda = 1, 2$). As a result explicit expressions of the soft-mode frequency Ω_0 at the X-point of BZ and of the order parameter $|\langle R_\lambda(\ell)\rangle|$ were given /6/.

To introduce the deformation we follow the notations of /7-10/ expanding the potential energy of the distorted lattice in terms of the localized strain tensor e_{ij} :

$$H_e = \frac{1}{2}\sum_{\ell,\alpha} M\dot{u}_\alpha^2(\ell) + \sum_\ell [\frac{1}{2}C_{11}\{e_{11}^2(\ell) + e_{22}^2(\ell)\} +$$

$$+\frac{1}{2}C_{33}e_{33}^2(\ell) + C_{13}\{e_{11}(\ell)e_{33}(\ell) + e_{22}(\ell)e_{33}(\ell)\} + \tag{3}$$

$$+C_{11}e_{11}(\ell)e_{22}(\ell) + \frac{1}{2}C_{66}e_{12}^2(\ell) + \frac{1}{2}C_{44}\{e_{12}^2(\ell) + e_{23}^2(\ell)\}].$$

C_{ij} are the elastic constants of the tetragonal lattice, M is the total mass of atoms in the unit cell. $u_\alpha(\ell)$ and $M\dot{u}_\alpha(\ell)$ are the position and momentum of the c.m. of the l-th unit cell, respectively. The elastic strains can be expressed by means of the normal coordinates $Q(\mu, \vec{q})$ of the μ-th acoustic branch with frequency $\omega(\mu, \vec{q})$, wave vector $\vec{q}$ and polarization vector $\vec{e}(\mu, \vec{q})$:

$$e_{ij}(\ell) = \langle e_{ij}(\ell)\rangle + u_{ij}(\ell), \tag{4-1}$$

$$u_{ij}(\ell) = \frac{i}{2\sqrt{N}}\sum_{\mu,\vec{q}} e^{i\vec{q}\vec{\ell}}[q_i e_j(\mu,\vec{q}) + q_j e_i(\mu,\vec{q})]Q(\mu,\vec{q}), \tag{4-2}$$

$$\langle e_{11}(\ell)\rangle = \epsilon_1, \quad \langle e_{22}(\ell)\rangle = \epsilon_2, \quad \langle e_{33}(\ell)\rangle = \epsilon_3, \quad \langle e_{12}(\ell)\rangle = \epsilon_6. \tag{4-3}$$

The static deformation ϵ_i is taken into consideration in the long-wave length limit. The dynamical part of (3) becomes

$$H_e^d = \frac{1}{2M}\sum_{q,\mu} P(\mu,-\vec{q})P(\mu,\vec{q}) + \frac{1}{2}M\sum_{\mu,\vec{q}}\omega^2(\mu,\vec{q})Q(\mu,\vec{q})Q(\mu,-\vec{q}). \tag{5}$$

The interaction between optical (soft) and acoustic phonons can be written as

$$H_{R-e} = \sum_{\substack{\alpha,\beta,\gamma,\phi \\ \ell,k,k'}} g_{\alpha\beta\gamma\phi}(k,k')\, e_{\alpha\beta}(\ell)\, X_{\gamma}(\ell,k)\, X_{\phi}(\ell,k') \, . \tag{6}$$

We are interested only in the coupling to the tilting mode:

$$H_{R-e} = \sum_{\substack{\alpha,\beta \\ \ell,k,k'}} g_{\alpha\beta}(k,k')\, e_{\alpha\beta}(\ell)\, X(\ell,k)\, X(\ell,k') \, . \tag{7}$$

Because of the tetragonal symmetry of the lattice for $T > T_0$ there exist only three independent components of $g_{\alpha\beta}$:

$$\alpha_0 = g_{xx}(k,k) = g_{yy}(k,k) \, ; \quad \beta_0 = g_{zz}(k,k) \, ; \quad \gamma_0 = g_{xy}(1,2) \, . \tag{8}$$

Transforming (7) with the help of (6) one gets:

$$H_{R-e} = \frac{1}{8m} \sum_{\substack{\ell,\ell' \\ \lambda,\lambda'}} W_{\lambda\lambda'}(\ell)\, \sigma_{\lambda\lambda'}(\ell,\ell')\, R_{\lambda}(\ell)\, R_{\lambda'}(\ell') \, , \tag{9-1}$$

with

$$W_{\lambda\lambda'}(\ell) = \begin{cases} \alpha_0 \{ e_{xx}(\ell) + e_{yy}(\ell) \} + \beta_0 e_{zz}(\ell) \, , & \lambda = \lambda' \, , \\ \gamma_0 \, e_{xy}(\ell) \, , & \lambda \neq \lambda' \, , \end{cases} \tag{9-2}$$

$$\sigma_{\lambda\lambda'}(q) = \begin{cases} 2(1 - F_{\lambda}(q)) \, , & \lambda = \lambda' \, , \\ 1 - F_{\lambda}(q) - F_{\lambda'}(q) + F_{\lambda}(q)\, F_{\lambda'}(q) \, , & \lambda \neq \lambda' \, , \end{cases} \tag{9-3}$$

$$F_x(q) = \cos q_y a \, , \quad F_y(q) = \cos q_x (a) \, . \tag{9-4}$$

From eq. (9) one can obtain the correct $\epsilon \cdot R^2$ interaction in the phenomenological Landau expansion of the free energy /10/, where ϵ are components of stress tensor. Here we are interested only in the so-called resonant part of (9) producing a jump in the sound velocity at T_0 :

$$H_{res} = \frac{iR}{8m\sqrt{N}} \sum_{\mu,\vec{q}} Q(\mu,\vec{q}) \sum_{\ell,\lambda,\lambda'} M_{\lambda\lambda'}(\mu,\vec{q})\, \sigma_{\lambda\lambda'}(\vec{q}+\vec{q}_1)\, e^{i\vec{\ell}(\vec{q}+\vec{q}_1)} \cdot r_{\lambda}(\ell) \quad (10)$$

with

$$M_{\lambda\lambda'}(\vec{q},\mu) = \begin{cases} (a_0\vec{q}_{\perp} + \beta_0\vec{q}_z)\cdot\vec{e}(\mu,\vec{q}), & \lambda=\lambda', \\ \gamma_0\,(q_x e_y(\mu,q) + q_y e_x(\mu,q)), & \lambda\neq\lambda', \end{cases} \quad (11)$$

$R = |\langle R_{\lambda}(\ell)\rangle|, \quad r_{\lambda}(\ell) = R_{\lambda}(\ell) - \langle R_{\lambda}(\ell)\rangle.$

This resonant part vanishes in the tetragonal phase ($R = 0$). In the case of $T < T_0$ the equation of motion of the acoustic commutator Green's function $D(\mu, \vec{q}, \omega)$ becomes:

$$D(\mu,\vec{q},\omega) = [\,\omega^2 - \omega^2(\mu,\vec{q}) - \Sigma(\mu,\vec{q},\omega)\,]^{-1}. \quad (12)$$

Neglecting the terms $\langle Q(\mu,\vec{q})\, r_{\lambda}(\ell)\rangle$, the mass operator Σ can be expressed by the commutator Green's function $G_{\lambda\lambda'}(\vec{q},\omega)$ of the tilting mode:

$$\Sigma(\mu,\vec{q},\omega) = \frac{4R^2}{(16m)^2}[\sum_{\lambda'} M_{\lambda\lambda'}(\mu,\vec{q})\,\sigma_{\lambda\lambda'}(\vec{q}-\vec{q}_1)]^2 \cdot \sum_{\lambda\lambda'} G_{\lambda\lambda'}(\vec{q}-\vec{q}_1,\omega). \quad (13)$$

In the long-wave length limit of acoustic phonons [1-4] we can approximate $\vec{q}-\vec{q}_1 \approx -\vec{q}_1$.

The corresponding acoustic frequencies satisfy the inequality $\omega(\mu,\vec{q}) \ll \Omega_0(T)$. Using

$$G_{\lambda\lambda'}(\vec{q}_1,\omega) = [\,-\Omega_0^2(T) + \omega^2\,]^{-1} \cdot \delta_{\lambda\lambda'},$$

(see [6]) one can calculate the new frequency poles of $D(\mu, \vec{q}, \omega)$ and consequently the renormalized frequencies. The lower frequency corresponding to the modified acoustic energy is

$$\tilde{\omega}^2(\mu,\vec{q}) \approx \omega^2(\mu,\vec{q}) - \frac{a_{\mu}(\vec{q})}{\Omega_0^2}, \quad (14)$$

with

$$a_{\mu}(\vec{q}) = \frac{8R^2}{(16m)^2}[\sum_{\lambda'} M_{\lambda\lambda'}(\mu,\vec{q})\,\sigma_{\lambda\lambda'}(\vec{q}_1)]^2.$$

To compare (14) with the experiments [1-4] one has to integrate $\alpha_\mu(\vec{q})$ over all $\vec{q}$-directions as soon as only ceramic samples were used. In the case of longitudinal sound it follows:

$$\alpha_L(q) \approx \frac{1}{2}\left(\frac{R}{3m}\right)^2 (2\alpha_0 + \beta_0)^2 q^2 , \tag{15-1}$$

and in the case of transverse sound:

$$\alpha_T(q) \approx \frac{1}{2}\left(\frac{R}{m}\right)^2 \gamma_0^2 q^2 . \tag{15-2}$$

Using $\Omega_0^2(T) = 32\Gamma_0 R^2(T)$ for $T < T_0$ (see [6]) one gets (even in the case of a second order SPT as in La_2CuO_4) for the changed sound velocity $\tilde{S}_\mu$:

$$\tilde{S}_L = [S_L^2 - \frac{1}{9}\frac{1}{B}(2\alpha_0 + \beta_0)^2]^{1/2} , \tag{16-1}$$

$$\tilde{S}_T = [S_T^2 - \frac{1}{B}\gamma_0^2]^{1/2} . \tag{16-2}$$

The change in S_μ is finite as predicted by experiments. The jump height is determined by the anharmonicity of the oxygen-Z-vibrations $B = 64\Gamma_0 \cdot m^2$ and the interaction parameters α_0, β_0 and γ_0 .

Let us discuss the influence of Sr-doping. Besides of changes in the electronic structure, there is a strong dependence of $S_\mu^2 - \tilde{S}_\mu^2$ on x_{Sr} with its maximum near $x = 0.15$. As was shown in [6], there will be no noticeable B(x)-dependence, but according to (16) an increase of γ_0 with x_{Sr} up to $x \approx 0.15$ and its drastic decrease for $x > 0.15$ is to be expected. Together with the fact that S_T (for $T > T_0$) has its minimum at $x \approx 0.2$ we conclude the Sr-doping causes (besides a decrease of T_0 — see [6]) a remarcable softening of the elastic constants $C_{ij} \sim S_\mu$ in the tetragonal phase (up to $x \approx 0.2$) and furthermore an increase of the interaction strength between the deformation and the optical soft mode (up to $x = 0.15$). In summary these effects lead to a softening of the elastic constants of 50% at low temperatures for $0.15 < x < 0.2$. Thus, the possible role of the SPT in $La_{2-x}Sr_xCuO_4$ is to cause a softening of the lattice in the orthorombic phase. Note, that the change in the sound velocity (16) is independent of temperature. This is also an unexplained experimental fact [1-4] suggesting our model to be valid over a wide temperature range. Another interesting fact is the wide temperature range of 60K-150K, where the change

$S_T \to \tilde{S}_T$ takes place. This may be caused by additional slow-relaxation dynamics. As was mentioned in [6], there are indications of nonvanishing long-time correlations $L_{\lambda\lambda} = \lim_{t \to \infty} \langle r_\lambda(t) r_\lambda \rangle \neq 0$ near the SPT, causing central peak and precursor cluster fluctuations (see [12]). It is easy to show in the case of $L \neq 0$ that there exists a resonant part in (9) even for $T > T_0$, which has the same form as (10) where the order parameter R^2 is replaced by L.

Summarizing, a model for the microscopic description of anomalous lattice behaviour in $La_{2-x}Sr_xCuO_4$ is presented. The SPT can "switch on" a remarcable softening of the elastic moduli. This softening may change through crystal fields inner properties of some excitations in the crystal, e.g. the frequency of excitons connected with d-d-excitations (see [13]). Thus, the interaction of such excitations with electrons is changed.

References

1. Horie Y., Fukami T., Mase S. — Solid State Comm., 1987, 63,p.653.
2. Bishop D.J., Gammel P.L. — Phys.Rev., 1987, B35, p.8788.
3. Fossheim K., Laegrid T. et al. -- Solid State Comm., 1987, 63, p.531.
4. Yoshizaki R., Hikata T. et al. -- J.J.Appl.Phys., 1987, 26, Suppl.26-3.
5. Birgeneau R.J., Chen C.Y. et al. — Phys.Rev.Lett., 1987, 59, p.1329.
6. Aksenov V.L., Flach S., Plakida N.M. — phys.stat.sol.(b), 1988, 146.
7. Feder J., Pytte E. -- Phys.Rev., 1970, B1, p.4803.
8. Pytte E. — Phys.Rev., 1970, B1, p.924.
9. Konwent H., Plakida N.M. -- Acta Phys.Pol., 1983, A63, p.755.
10. Plakida N.M., Shakhmatov V.S. — Physica C, 1988, C153 in print.
11. Fossheim K. Physica Scripta, 1982, 25, p.665.
12. Aksenov V.L., Bobeth M., Plakida N.M., Schreiber J. — J.Phys., 1987, C20, p.375.
13. Weber W. -- J.Phys. B., Z.Phys., 1988, 70, p.323.

Received on May 6, 1988.