THE A_{1g} MODE IN THE Hg-1201 PHONON SPECTRUM AS AN INDICATOR OF N→S TRANSITION

YA. DOVHYJ

Ivan Franko National University of Lviv
(8, Kyrylo–Mefodiy Str., Lviv 79005, Ukraine; e-mail: dovgy@physics.wups.lviv.ua)

PACS 74.62.Fj, 74.25.Gz, 74.25.Kc

By analyzing the structure of and the temperature changes in $HgBa_2CuO_{4+y}$ phonon spectra, the electron-phonon coupling constant g has been determined for the first time. It is shown that this compound is a superconductor with strong coupling. A frequency interval around 60.4 meV in the $HgBa_2CuO_{4+y}$ phonon spectrum, which may be classed as a "soft mode", is revealed. The dominant partial contribution to the density of phonon states in that spectral range is found to be given by O(2) atomic vibrations.

The fact that mercury-based high-temperature superconductors are characterized by a high baric coefficient $\beta = dT_c/dP$ means that local lattice deformations can be more important here than in other high-temperature superconductors (HTSCs) [1]. Therefore, careful researches of the lattice dynamics in Hg-based HTSCs became the subject of a special attention for experimenters. In addition to the traditional optical spectral researches of phonon spectra, x-ray absorption spectroscopy and neutron scattering are most often in use.

In this work, the magnitude of electron-phonon coupling constant in the high-temperature superconductor $HgBa_2CuO_{4+y}$ (Hg-1201) is determined for the first time. The authors of work [2] considered the fine structure of the x-ray absorption spectra (XAFS spectra) of $HgBa_2CuO_{4+y}$ with $T_c = 96$ K in order to reveal dynamic correlations (or anticorrelations) at Cu–O(2) and Ba–O(2) bonds (see Fig. 1). The analysis of this structure showed that, at $T = 80$ K, the relative displacements of Cu atoms and axial O(2) atoms, as well as Ba and O(2) atoms, are anticorrelated. This means that the correlation parameter Φ in the expression

$$\sigma_{ab}^2 = \sigma_a^2 - \sigma_b^2 - 2\sigma_a\sigma_b\Phi \qquad (1)$$

for the root-mean-square deviation of atoms a and b is negative ($\Phi < 0$). The case $\Phi = 1$ corresponds to the strong correlation between neighbor atoms, whereas the case $\Phi = 0$ to the non-correlated dynamics of more remote atoms.

Which is the reason for the anticorrelation at the Cu–O(2) and Ba–O(2) bonds? It may possibly be associated with the fact that the position of O(2) atom can vary, by depending on whether the neighboring O(3) position is occupied or not. Really, the studies of $HgBa_2CuO_{4+y}$

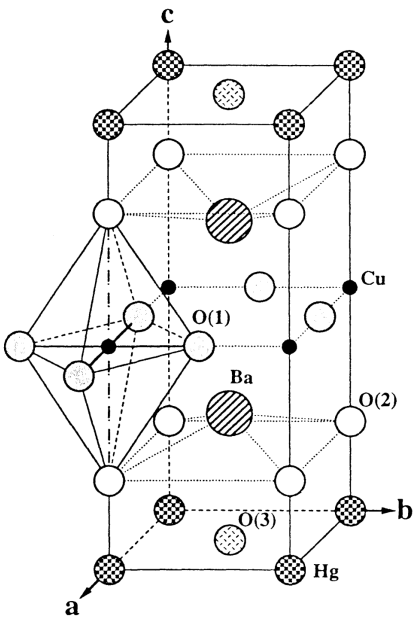

Fig. 1. Crystal structure of $HgBa_2CuO_{4+y}$ at room temperature

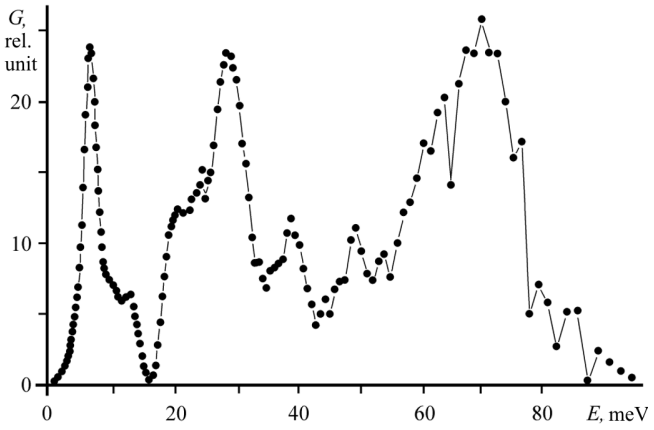

Fig. 2. Phonon density of states in $HgBa_2CuO_{4+y}$. O(3) positions correspond to oxygen vacancies

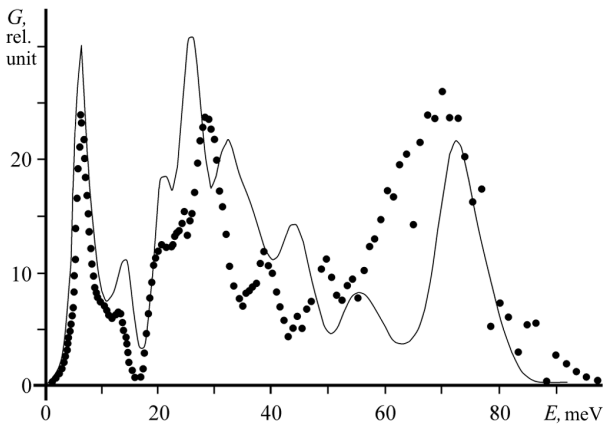

Fig. 3. Comparison between the experimental spectrum (points) and the results of model calculations (solid curve)

Raman spectra showed that the A_{1g} mode consists of two components, which follows from the peculiarity of O(2) atomic vibrations. The main peak in the A_{1g} doublet corresponds to 592 cm^{-1}, and the less intensive one to 570 cm^{-1} [3]. Hence, we have two possible positions of O(2) atoms, which is connected with a local redistribution of charge. This factor may also be important for the mechanism of charge carrier pairing in CuO_2 layers. By the way, as was found in work [4], the A_{1g} doublet is very sensitive to the pressure.

On the basis of researches using the inelastic scattering of low-energy neutrons, the generalized density of states in $HgBa_2CuO_{4+y}$ (Fig. 2) was successfully obtained [5]. Measurements were carried out on a Grenoble reactor. A monoenergetic beam of 17.4-meV neutrons was used. The scattering intensity was registered in the angular range of $23° < \alpha < 99°$, which, together with the known processing technique of results, provided the monitoring over a considerable volume in the momentum space and the determination of reliable values for the density of phonon states $G(E)$.

The high-energy section in the spectrum structure $G(E)$ can be associated with oxygen modes, similarly to what takes place in $YBa_2Cu_3O_{7-\delta}$ and Bi-based HTSCs [6]. In particular, the main peaks at 73.4 meV (592 cm^{-1}) and 20.8 meV (168 cm^{-1}) correspond to the A_{1g} and E_g modes of the O(2) vibrational spectrum.

In Fig. 3, the experimental spectrum $G(E)$ is confronted with the results of model calculations. The force constants and other calculation parameters were taken from review [7], in which a large body of experimental data concerning superconducting cuprates was analyzed. The figure demonstrates that the experimental and calculated spectra $G(E)$ are in qualitative agreement. A divergence in the high-energy section can be explained by the influence of defects in real ceramic specimens. Well-pronounced peaks in the vicinity of 28 meV (226 cm^{-1}) and 70 meV (565 cm^{-1}) correspond to the TO and LO vibrational modes of vertex oxygen atoms. Note that those modes are characteristic of Hg-based HTSCs with various numbers of CuO_2 layers in the unit cell.

It is known [8,9] that the phonon mode with B_{1g} symmetry (335 cm^{-1}) in $YBa_2Cu_3O_{7-\delta}$ behaves as a "soft" one, being related to the existence of a superconducting gap 2Δ. Does a similar mode exist in Hg-based HTSCs? In this connection, it is of considerable interest to analyze the temperature behavior of the phonon spectra in those substances.

In Fig. 4, the temperature-induced variations in the $G(E)$ function are depicted. They were obtained using the inelastic neutron scattering in $HgBa_2CuO_{4+y}$ specimens [5]. One can see that substantial modifications of the spectrum are observed in the vicinity of 60.4 meV, when the specimen is cooled down to the temperature of the transition into the superconducting state. In other sections, the spectrum does not undergo appreciable temperature-induced changes.

Hence, the 60.4-meV mode in $HgBa_2CuO_{4+y}$ reveals certain attributes of "soft mode". The temperature-induced variations testify that the largest contribution to $G(E)$ in this frequency range is made by O(2) vibrations, and the given mode can be related to a transition from the normal into the superconducting phase. This assumption was confirmed in work [10], where the authors observed similar temperature-induced variations in the Raman spectra of Hg-based HTSCs. A soft vibration mode for the Cu–O(2) bond was also found by

the authors of work [11], while studying optimally doped $HgBa_2CuO_{4+y}$ specimens.

By analogy with $YBa_2Cu_3O_{7-\delta}$, we suppose that the energy gap in $HgBa_2CuO_{4+y}$ is

$$2\Delta \cong 60.4 \text{ meV}, \tag{2}$$

whence

$$2\Delta/(kT_c) \approx 7.2. \tag{3}$$

In the Bardeen–Cooper–Schrieffer (BCS) theory, the ratio $2\Delta/(kT_c) = 3.52$, which is half as large as in Eq. (3). Such an amazing discrepancy can testify, first of all, that the electron-phonon coupling in Hg-based HTSCs is much stronger than that in the BCS theory.

Data concerning the symmetry and the shape (in the momentum space) of the energy gap in a superconductor on the Fermi surface are known to be rather important for the analysis of pairing models. In Hg-1201, as well as in other cuprates, superconductivity is governed by the d-type Cooper pairing, so that the gap can possess nodes on the Fermi surface. However, the absence of detailed data on the Fermi surface topology does not allow one to determine a character of the d-pairing anisotropy in the given substance.

The temperature behavior of the "indicator band" testifies to a substantial role of the electron-phonon interaction in the pairing. The spin-fluctuation mechanism cannot be excluded from consideration as well, because cuprates were reliably established to be systems with strong electron correlations. Other models [12] are also noteworthy. In this connection, an unequivocal interpretation of the nature of the energy gap in Hg-1201 is problematic.[1]

In this work, an attempt was made to evaluate the constant of electron-phonon coupling g in the framework of strong-coupling model. We used the Allen–Dynes approximation formula [14]

$$T_c \cong \left(\frac{f_1 f_2}{1.20}\omega_a \frac{\hbar}{k}\right) \exp\left[-\frac{1.04(1+g)}{g-\mu^*(1+0.62g)}\right], \tag{4}$$

where

$$f_1 = \left[1 + (g/a_1)^{3/2}\right]^{1/3}, \tag{5}$$

$$f_2 = 1 - \frac{g^2\,(1-\omega_0/\omega_a)}{g^2 + a_2^2}, \tag{6}$$

[1] Note that, for systems with the partial gapping of the electron spectrum, the emergence of a Mott-type gap is also probable [13].

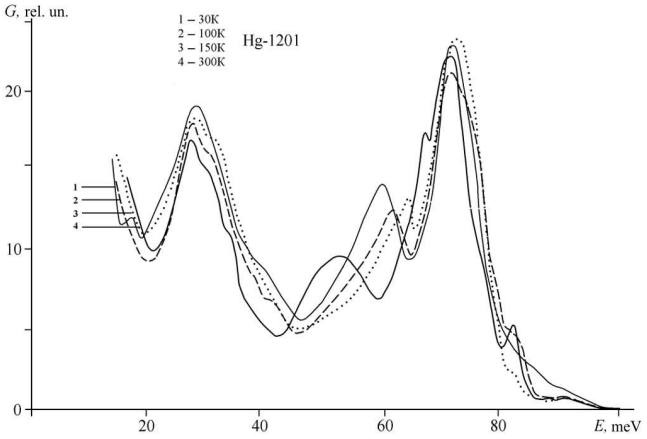

Fig. 4. Temperature-induced changes of the phonon modes for $HgBa_2CuO_{4+y}$

$$a_1 = 2.46(1 + 3.8\mu^*), \tag{7}$$

$$a_2 = 1.82(1 + 6.3\mu^*)\omega_0/\omega_a, \tag{8}$$

$$\mu^* = \frac{\mu}{1 + \mu \ln\left(E_{\rm F}/(\hbar\omega_{\rm D})\right)}, \tag{9}$$

μ is a parameter that characterizes the Coulomb repulsion between carriers, $\omega_{\rm D}$ is the Debye frequency, and k is the Boltzmann constant. From the analysis of the function $G(E)$ represented in Fig. 2, we obtain

$$\hbar\omega_0 \cong 28.5 \text{ meV}. \tag{10}$$

Taking into account that

$$\hbar\omega_a \cong 2\Delta, \tag{11}$$

and, according to work [15], the parameter of the modified Coulomb potential is

$$\mu^* \cong 0.13, \tag{12}$$

we obtain after cumbersome calculations that

$$g \cong 2.0. \tag{13}$$

Therefore, we may assert that the mercury-based high-temperature superconductors are characterized by a strong electron-phonon coupling. The search for regularities in variations of the parameter g depending on the number of CuO_2 layers in a unit cell will be a subject of our subsequent researches.

1. Ya.O. Dovhyi, Ukr. Fiz. Zh. **46**, 585 (2001).

2. C.H. Booth, F. Bridges, E.D. Bauer, G.G. Li, J.B. Boyce, T. Claeson, C.W. Chu, and Q. Xiong, Phys. Rev. B **52**, 15745 (1995).
3. H.G. Lee, H.S. Shin, I.S. Yang, J.J. Yu, and N.H. Hur, Physica C **233**, 35 (1994).
4. I.-S. Yang, H.-S. Shin, and H.-G. Lee, Phys. Rev. B **51**, 644 (1995).
5. B. Renker, H. Schober, and F. Gompf, J. Low. Temp. Phys. **105**, 843 (1996).
6. B. Renker, F. Gompf, D. Ewert, P. Adelmann, H. Schmidt, E. Gering, and H. Mutka, Z. Phys. B **77**, 65 (1989).
7. S.L. Chaplot, W. Reichardt, L. Pintschovius, and N. Pyka, Phys. Rev. B **52**, 7230 (1995).
8. *Physical Properties of High Temperature Superconductors*, edited by D.M. Ginsberg (World Scientific, Singapore, 1989).
9. R. Zeyher and G. Zwicknagl, Z. Phys. B **78**, 175 (1990).
10. M.C. Krantz, C. Thomsen, Hj. Mattausch, and M. Cardona, Phys. Rev. B **50**, 1165 (1994).
11. H. Uchiyama, A.Q.R. Baron, S. Tsutsui, Y. Tanaka, W.-Z. Hu, A. Yamamoto, S. Tajima, and Y. Endoh, Phys. Rev. Lett. **92**, 197005 (2004).
12. Ya.O. Dovhyi, *A Miracle Phenomenon of Superconductivity* (Yevrosvit, Lviv, 2000) (in Ukrainian).
13. A.M. Gabovich, A.I. Voitenko, and M. Ausloos, Phys. Rep. **367**, 583 (2002).
14. P.B. Allen and R.C. Dynes, Phys. Rev. B **12**, 905 (1975).
15. H.B. Radousky, T. Jarlborg, G.S. Knapp, and A.J. Freeman, Phys. Rev. B **26**, 1208 (1982).

Received 08.06.11.
Translated from Ukrainian by O.I. Voitenko

A_{1g}-МОДА В Hg-1201 ЯК ІНДИКАТОР N→S ПЕРЕХОДУ

Я. Довгий

Р е з ю м е

На основі аналізу структури і температурних змін фононних спектрів $HgBa_2CuO_{4+y}$ (Hg-1201) вперше визначено константу електрон-фононної взаємодії g і показано, що Hg-ВТНП є надпровідниками з сильним зв'язком. У фононному спектрі $HgBa_2CuO_{4+y}$ виявлено ділянку частот поблизу 60,4 меВ, що має ознаки "м'якої моди". Основний парціальний внесок у функцію густини фононних станів у даній ділянці спектра дають коливання атомів O(2).