

- · · ·

Объединенный институт ядерных исследований

дубна

E17-88-382 e

T.Galbaatar,¹ R-J.Rakauskas,² J.Šulskus³

NUMERICAL SIMULATION OF THE ISOTOPE EFFECT IN THE HIGH-T_c SUPERCONDUCTORS

Submitted to "International Journal of Modern Physics B"

¹ On leave from Institute of Physics & Technology, Ulan-Bator-51, Mongolia
² On leave from Vilnius V.Kapsukas University, Faculty of Physics, 232054, Vilnius, USSR
³ Vilnius V.Kapsukas University, Faculty of Physics, 232054, Vilnius, USSR.

1988

I. INTRODUCTION

The recently synthesized cuprate oxide superconductors having transition temperatures T up to 114K $^{\prime1,2,3\prime}$ are very interesting research objects both from theoretical and experimental aspects. A major attention herein has been paid to the mechanism(s) of the formation of the superconducting state, in particular, whether it is mediated by phonons or not. It is well known that within the BCS theory the phonon mechanism is supported by the experimental observation of the effect of isotope substitution on the transition temperature T,. The mass dependence of T is expressed through T_~ $M^{-\alpha}$, where M is the isotopic mass and α is usually around 0.5. Thus, in the light of the new high-T superconductors the isotope effect becomes an important issue with respect to testing any theory intending to explain such high T_'s. Recently, an oxygen isotope effect in both the LaSrCuO and YBaCuO systems has been observed ^{74,57}. Although the observed shifts in T were smaller than one would expect from the BCS theory,(9.3-1)K for LaBroud and (0.3 0.5)K for YBacuo, they nevertheless indicate that phonons are yet involved in superconductivity. Structural studies of the high-T cuprate oxides have led to the identification of La Ba CuO as an analogy of K_{2}^{NiF} and z-x x 4 z 4 be a previously unknown oxygen-deficient YBa Cu O to perovskite²⁸.Evidence of structural instabilities and the existence of soft modes in LaSrCuD system have been reported $^{2-12}$. The study of the phonon behaviour of YBa Cu 0 by neutron inelastic scattering show larger phonon density states in lower frequency modes rather than in high frequency ones¹⁸⁷. Another interesting feature, namely a possible softening of the high frequency breathing mode was observed ^{/14/} in YBa_Cu_O_.Thus, these two types of the oxygen vibration, e.g., the high frequency in-plane bond-stretching mode and the low frequency out-of-plane one, the so-called bond-bending mode are of interest within the conventional electron-phonon mechanism. As superconductivity is observed in ceramic and crystalline states, it is practical to apply the cluster approximation method in the theoretical consideration of superconductivity in these compounds. On the

Возсанов иный институт па-уних иссоразная

other hand taking into account the sufficient difference in the masses of oxygen and copper. one can make use of a guasi-local mode describing the vibration of the oxygen ion, only. Calculations of the electronic structure^{/13/} demonstrate overlapping of the 3d states of copper with the 2p states of oxygen resulting in strong coovalency which in its turn might cause a strong anharmonicity for the oxygen vibration. Therefore, by considering the electron-phonon interaction in the new high-T compounds it is important to take into account the effect of anharmonicity.

II. MODEL OF THE ANHARMONIC QUASI-LOCAL MODE

We suggest for the vibration of the oxygen ions a double-well anharmonic model potential

 $U_{(x)} = -Ax^{2}/2 + Bx^{4} + A^{2}/4B, \qquad (1)$ where A and B are positive and related through $U_{0} = A^{2}/4B \quad \text{and} \quad x_{0} = (A/B)^{1/2}$ to real physical quantities, the central barrier height and it's

average width, respectively, thus $2x_{n}$ being the distance between the two minima of the potential.Units are used where h=k_=1. As a mailer of fact, several authors '6,15,16' have been using the "Gaussian barrier" type double-well potential function, superposed of a Gaussian and a harmonic oscillator potential function.

Although this construction merits attention from a perturbative theoretical point of view, for a good approximation to it the knowledge of three independent properties is required, whereas for the potential (1) only one free parameter

$$\beta = \omega_{\circ}/4U_{\circ}, \qquad (2)$$

where $\omega = \sqrt{\frac{2A}{m}}$ is a characteristic frequency; and m, the reduced mass of the Cu-O-Cu cluster, thus a complete substitution of by 0^{10} in the cluster is simulated through decreasing β by 5%. In this respect, it is rather advantageous to work with the ootential function (1).

been used to On the other hand (1) is more physical and has describe vibrational effects. Here, we recall the work explaining the infrared spectra of H-bonded solids. Moreover it should be pointed out that this potential was used to describe

structural phase transitions of order-disorder or displacive types 96 . Indeed, if the central barrier height U \gg T a transition due to ordering takes place, whereas if $U_{x} \propto T_{z}$ a displacement of atoms between the two minima occurs. This fact and the observation of structural instabilities and soft modes in the cuprate oxides provide a reasonable background for applying this potential to the new superconducting materials.

By introducing the dimensionless coordinate and energy

$$\xi = x/x_o$$
 and $E_n = E'_n / U_o$ (3)
the Schrödinger equation

 $-(1/2m)d^2 \psi(x)/dx^2 + U(x)\psi(x) = E'_{\mu}\psi(x)$ (4)is converted into the following form

$$-d^{2}\varphi(\xi)/d\xi^{2} + 1/\beta^{2}[(1-\xi^{2})^{2} - E_{n}]\varphi(\xi) = 0, \quad (5)$$

which was numerically solved for a broad set of the dimensionless parameter β , yielding the energy spectrum E_n of the anharmonic oscillator. For the sake of certainty of the results we carried out the calculations by two different methods:

(i) A finite difference approach substituting the differential operator by a series of the central difference operator up to the sixth power, thus transforming the Schrödinger equation into an algebraic eigenvalue problem /18,19/:

(ii)Direct numerical integration and shooting method ^{20,21/}.

The sufficient reproducibility of the results was demonstrated by the coincidence of the eigenvalues to the 8th figure.The anharmonic eigenfunctions $\varphi(x)$ are expanded in terms of the eigenfunctions u_i(x) of the harmonic oscillator

$$\varphi_{n}(x) = \sum_{j=0}^{N_{max}} a_{j} u_{j}(x) , \qquad (6)$$

where Nmax is fixed by the condition that $\sum_{j=0}^{Nmax} a^2_j$ be unity within a given accuracy. A study on the convergence accuracy of (6) indicates that it improves from 0.25% to better than 0.0001% when the number of the expansion coefficients is increased from 40 to 100. This demonstrates the strong effect of the anharmonicity of the eigenfunctions and difficulties related to a perturbative treatment of the problem.

In FIG.1 the vibrational frequencies $oldsymbol{arepsilon}_{i,i}$ given by

$$\alpha_{ij} = \varepsilon_{ij} U_o = (\varepsilon_j - \varepsilon_i) U_o \tag{7}$$

are plotted vs. β .For large heights of the central barrier, e.g. for small β 's the transitions are due to tunnelling. Comparing our results with those obtained within the WKB approximation (dashed line in FIG.1)²²⁷ we found a good agreement for ε_{01} up to the value β =0.1195 while for higher levels a significant deviation is observed as well as for ε_{01} at larger β' . The vibrational frequency tends to saturate with increasing β (smaller U_0), which means the oscillator seems not to feel the anharmonicity. 10





This behaviour of the energy spectrum is illustrated in FIG.2 in terms of

 $\Delta_{1+4,3+1} | \mathcal{E}_{1,3}$.



Here is

e

 $\Delta_{1+1,2+1} = E_{2+1} - E_{1+1} - E_{1+1}$

(8)

There, one can see two features of the above ratio, demonstrating strong for higher levels weak and for lower ones anharmonicity. This feature is also observed in the dependence of the mean square amplitude of vibration (Table 1) on the anharmonicity. For the mean square amplitude of vibration calculated by

$$Q^2 = \langle O | \xi^2 | 1 \rangle , \qquad (9)$$

we found that while for larger β it steadily increases with temperature, for lower values of β an oscillating behaviour is observed.

5

Table 1. The mean-square amplitude of vibration (dimensionless) for different values of β .Underneath the corresponding eigenvalues E_n (dimensionless) are given. n is the order number of the energy levels.

<u>o</u>	0	1	2	3	4	5	6	7	Θ.
.125	.951	.932	.753	.785	.452	.718	.717	.815	.897 1 77
tn .42	.242	.243 .922	.801	1.08	1.24	1.30	1.54	1.65	1.79
En .6	.631 .594	.842 1.01	1.79	2.71 1.31	3.04 1.53	5.12 1.69	6.51 1.89	8.00 2.04	2.27
En 84	.777	1.33	2.82	4.48	6.41 1.68	8.58 2.07	10.88	13.37	15.99 2.73
En	.982	2.12	4.51	7.26	10.41	13.89	17.64	21.64	25,85

III. EFFECT OF ANHARMONICITY ON ELECTRON-PHONON INTERACTION

In this section we apply the numerical results to calculate the transition temperature T_c and the isotopic shift δT_c of the high-Tc superconductors within a conventional phonon mediated mechanism for the attractive electron-electron interaction. In fact, it was shown 237 that transition temperatures up to 40K can be explained within the phonon mechanism. Moreover, values of Tc as high as 90K are possible 247 provided one gets for the electron-phonon coupling values around 2.However, the pure phonon mechanism in the harmonic approximation fails to explain the experimentally observed 90K in the YBaCuO compound 247 .

Dur starting point to study the anharmonic effect on the electron-phonon interaction is the anharmonic model of high-Tc superconductors proposed in $^{\prime 25\prime}$. In this paper, within a two-level approximation by applying the two-time Green function technique the Eliashberg equation and the following relation for the coupling constant $\lambda_{\rm a}$ were obtained

$$\lambda_c = N(o) < J_s^2 > X_s, \qquad (1)$$

where Noo, J and χ_{g} denote the density of states at Fermi level E_g, the deformation potential and the static susceptibility of the lattice, respectively. The latter is described in a two-level approximation by

$$\pi_{s} = \frac{2x_{o}^{2} \xi_{o1}^{2}}{R_{o1}} \ln(R_{o1}/2T), \qquad (14)$$

where $x_{0i} = x \xi_{0i}$ is the matrix element of the dipole moment between the states φ_0 and φ_i . Expression (11) can be derived from the spectral representation

$$\chi_{s} = Z^{-1} \sum_{n,m} e^{-\overline{T} E_{n}} \frac{4 - e^{-\overline{T} (E_{m} - E_{n})}}{(E_{m} - E_{n})/U_{o} - \omega/U_{o}} < m |\xi|^{n} >^{2}, \quad (42)$$

where $Z=\sum e^{-En/T}$ in the static limit $\omega =0$ by considering the two lowest levels,only $^{/39/}$. Thus, in accordance with the energetic criterion for the application of the two-level model given in FIG.2 we consider the case $\beta \otimes .9$, because namely in that region the two lowest levels are separated from the higher levels by a large energy gap relative to the energy difference $(E_i - E_o)$. The lower limit of β can be estimated by the relation (7). We reckon the frequency Ω_i , in the case of the soft bond bending mode at range 10-30meV 734 , 35/. Then, for values of β -0.2 U₀ becomes very large (~1000 meV) which is unphysical. In contrast with it in the region β =0.4+0.8 U₀ takes values between 10-150meV, thus values of β in this range can be considered as physically meaningful.

The transition temperature has been calculated by the general relation

$$T_{c} = \Omega_{o_{A}} f(\lambda_{s}, \mu^{*}), \qquad (43)$$

where Ω_{01} is the frequency of the phonon mode under consideration, $f(\lambda_{\bullet}, \mu^{\bullet})$ expresses the dependence of T_c on the coupling constant and the Coulomb pseudopotential and is given for the weak and strong coupling cases by the McMillan ⁽²⁶⁾ and the Allan-Dynes ⁽²⁷⁾ formulas, respectively.

$$f(\lambda_{s}, \mu^{*}) = 0,83 \exp(-1,04(1+\lambda_{s})/g) \quad \text{for } \lambda_{s} < 1, \qquad (14)$$

$$f(\lambda_{s}, \mu^{*}) = 0,83(1 + (\lambda_{s}/\Lambda_{s})^{3/2})^{1/3} \exp(-1,04(1+\lambda_{s})/g) \quad \text{for } \lambda_{s} > 1, (15)$$
where
$$\Lambda_{s} = 2,46(1+3,8\mu^{*}), \qquad g = (\lambda_{s} - \mu^{*} - 0,62\lambda_{s}\mu^{*})/1,04.$$

Differentiating (n_{2}) with respect to the isotopic mass one obtains the following relation for the relative shift in T_

$$\delta T_c = dT_c / T_c = -\lambda dm / m, \qquad (18)$$

where

$$\mathcal{L} = 0,5\left(1 + \frac{B}{\varepsilon}\frac{d\varepsilon}{d\beta}\right)\left(1 - \frac{\lambda}{f}\frac{df}{d\lambda} - \frac{\mu^{2}}{f}\frac{df}{d\mu^{2}}\right). \quad (17)$$

The isotope effect thus depends on λ_{\bullet} and μ^{\bullet} through the second term while the first term gives the effect of the anharmonicity on

the value of α . The modification of the frequency $\Delta \varepsilon_i$ due to the variation $\Delta \beta$ is plotted in FIG.3 showing a saturation for the weak and almost vanishing for the strongly anharmonic case.

To calculate the coupling constant we estimate x_0 , No and J_a^2 in following ranges /28-90/:



FIG.3. The change of the vibrational frequency due to variation of β . 1- $\epsilon_{0,1}$; 2- $\epsilon_{2,3}$; $3-\epsilon_{4,3}$;

As for $\Omega_{\rm cal}$ the choice of the mode at present cannot take place unambiguously. Actually the high-frequency bond-stretching mode has so far been considered by many authors ^{/91,23,24/},to be responsible for high T's. Nevertheless,bond-bending-type interactions are able to produce large coupling constants because of the high polarizability ^{/24,30/}.This idea has been backed up by the observation of unusual large Debye-Waller factors in YBa_Cu_O /28,32/.By an appropriate combination of the structural and electronic data it is possible to obtain transition temperatures in the region of 90K, even for the soft bond-bending mode, the frequency of which is assumed to be in the range 10-30 meV /29,24,95/ within the anharmonic model.Concerning the isotope effect we state that it is much more complicated than in the harmonic limit. As is shown in FIG.4 even an inverse isotope effect is possible for T_ $\times33K$, while in the narrow region 34-35K $\,$ a nearly zero or heavily suppressed one is observed.At T_c slightly above 36K α lies in the range 0.1-0.2, depending on β ; thus, it is in agreement with experimental observation for the LaSrCuO system.However, for higher T_c we found normal isotope effect with a trend of α to increase with temperature and then to saturate at 0.3-0.4, which is in contradiction with the up-to-date observed trend of α to decrease³⁴⁴.



FIG.4. The dependence of the constant of the isotope effect α on the electron-phonon coupling. a) $\beta=0.84$; b) $\beta=0.4$; $1-\mu^{\bullet}=0$; $2-\mu^{\bullet}=0.1$; $3-\mu^{\bullet}=0.2$; The arrows -values of α at $T_{\alpha}=36K$.

We conclude from our numerical calculation that in the anharmonic model the phonon mechanism permits one to obtain an agreement with the experimentally observed isotope effect in the LaSrCuD system (a ~ 0.1-0.2) whereas it is not the case for the YBaCuO system. This may be an indication that higher energy levels have to be taken into account the more so as we have found an oscillating behaviour of the mean square amplitude of vibration(Table 1),otherwise a nonphonon mechanism should be considered. It is interesting to note that there is a qualitative agreement between our numerical calculations and the estimation of the isotope effect with the "Gaussian barrier" type potential '33'.Our main conclusion is that the two-level approximation is applicable in that region of β where the energy spectrum of the anharmonic oscillator has the feature that E is under the central barrier while the next level E is above it confirming the suggestion for the applicability of a two-level model '38'. This is consistent with the behaviour of the static susceptibility χ . We studied the convergence of (12) taking into account up to four levels. As one can see from Table 2. it is convergent,thus the application of the two-level approximation is justified. A more detailed study of the tunneling effect in a double-well potential e.g., region with small β was reported elsewhere 37 .

Table 2.The static susceptibility χ (dimensionless) calculated by taking into account 2,3 and 4 levels, respectively for three different values of β at T=92K.

ß		×,	
	<u>n</u> =2	n=3	<u>n=4</u>
.2	262.191	262.199	262.208
.5	3.236	3.249	3.251
1.0	.711	.719	.720

Finally, we would like to express our gratitude to Dr.N.M.Plakida for pointing out the problem and helpful discussions and Prof.M.G.Meshcheryakov for supporting the work.

References

.

1, Bednorz J.G. and Müller K.A. Z.Phys.B64,189(1986)

2, Wu M.K.et.al.	Phys.Rev.Lett.58, 908(1987)
3, Chu C.W. et.al.	Phys.Rev.Lett.60,941(1988)
4, Zur Loye H.C. et al.	Phys.Rev.Lett., 59, 915 (1987)
5, Learey K.J. et.al.	Phys.Rev.Lett.59, 1236(1987)
6, Clayman B.P. et.al.	Phys.Rev.B3 ,1351(1971)
7. Jorgensen J.D. et.al.	Phys.Rev.Lett.58, 1024(1987)
9, Liu J.O. et al.	Intern.J.Mod.Phys.B1,513(1987)
9, Cava R.J. et.al.	Phys.Rev.B35, 6716(1987)
10. Flemming R.M. et al.	Phys.Rev.B35, 7191(1987)
11. McPaul D. et al.	Phys.Rev.Lett.58, 1976(1987)
12. Hewat A.W. et al.	Nature, 327(1987)
13. Mattheiss L.F.	Phys.Rev.Lett.58, 1028(1987)
14. Renker B. et al.	2.Phys.B67, 15(1987)
15. Coon J.E. et al.	J.Molecul.Spectr. 20, 107(1966)
16, Baatar D., Plakida N.M	and Puzynin I.V.

JINR preprint 11-81-252, Dubna, 1981

17. Somorjai R.I. and Hornig D.F. J.Chem.Phys. 36, 1980(1962) 18. Fack V. and Van den BergheG.J.Phys.A18, 3355(1985) 19. Fack V. and Van den BergheG. CPC 19, 187(1986) 20, Cooley J.W. Math.commun. 15, 363(1961) 21, Johnson B.R. J.Chem.Phys. 67, 4086(1977) 22. Krumhansl J.A. and Schrieffer J.R. Phys.Rev.B11, 3535(1975) 23. Weber W. Phys.Rev.Lett.58, 1371(1987) 24. Weber W., Mattheiss L.F. Phys.Rev.B37, 599(1988) 25. Plakida N.M. et.al. Intern.J.Mod.Phys.B1,1071(1987) 26. McMillan W.L. Phys.Rev.167, 331(1968) 27. Allan P.B.and Dynes R.C. Phys.Rev.B12, 905(1975) 28. Capponi J.J.et al. Sol.st.comm.to be published 29. Benneman K.H. Phys.Lett.A,126, 67(1987) 30. Plakida N.M et.al. Europhys.lett.4, 1309(1987) 31, Batlogg B. et.al. Phys.Rev.Lett.58, 2333(1987) 32. Caponi J.J. et.al. Europhys.Lett. 3, 1301(1987) 33. Drechsler S.L.and Plakida N.M. phys.stat.sol.(b) 144, k113(1987). 34. Batlogg B. et.al. Phys.Rev.Lett.60, 754(1988) 35. Kulakovski V.D.et.al. Pisma v JETP 46, 460(1987) 36. Müller K.A.and Thomas H. Structural Phase Transitions I. lopics in current physics series Springer -Verlag,Berlin Heidelberg New York, 1981 37. Ionov S.P., et.al. Doklady akademi nauk USSR 193, 129(1970) 38. Vujičič G.M. et.al. Phys.Lett. 73A, 439 (1979)

> Received by Publishing Department on May 30, 1988.

10

39. Born M., Huang K. Dynamical Theory of Crystal Lattices

Clarendon Press, Oxford, 1954

SUBJECT CATEGORIES OF THE JINR PUBLICATIONS

Inde	c Subject
1.	High energy experimental physics
2.	High energy theoretical physics
3.	Low energy experimental physics
4.	Low energy theoretical physics
5.	Mathematics
6.	Nuclear spectroscopy and radiochemistry
7.	Heavy ion physics
8.	Cryogenics
9.	Accelerators
10.	Automatization of data processing
11.	Computing mathematics and technique
12.	Chemistry
13.	Experimental techniques and methods
14.	Solid state physics. Liquids
15.	Experimental physics of nuclear reactions at low energies
16.	Health physics. Shieldings
17.	Theory of condenced matter
18.	Applied researches
19.	Biophysics

Галбаатар Т., Ракаускас Р-И., Шулскус Ю.К. Численное моделирование изотопического эффекта в высокотемпературных сверхпроводниках

Представлены результаты моделирования кислородного изотопического эффекта в высокотемпературных сверхпроводниках, полученные численным решением уравнения Шредингера для квазилокальной моды движения атомов 01°, 01° с потенциалом вида $Ax^2/2 + Bx^4/4$. Обсуждаются особенности энергетического спектра, матричных элементов дипольного и квадрупольного операторов. Определена область применимости двухуровневой модели для описания электрон-фононного взаимодействия. В рамках данной модели изучено влияние изотопического замещения на T_c .

Работа выполнена в Лаборатории теоретической физики и Лаборатории вычислительной техники и автоматизации ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1988

Galbaatar T., Rakauskas R-J., Šulskus J. Numerical Simulation of the isotope Effect in the High-T_c Superconductors E17-88-382

The results of a numerical simulation are presented for the oxygen isotope effect in the high- T_c oxides by solving the Schrödinger equation for a local vibrational mode of oxygen ions in a double well anharmonic potential of the form $-Ax^2/2+Bx^4/4$. The features of the anharmonic energy spectrum, eigenfunctions and the matrix elements of the dipole and quadrupole moments are discussed. The applicability region of a two-level approximation to describe the electron-phonon interaction is given. The effect of isotope substitution on T_c in the new high- T_c superconductors is studied.

The investigation has been performed at the Laboratory of Theoretical Physics and the Laboratory of Computing Techniques and Automation, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1988

E17-88-382