LBL-31865

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

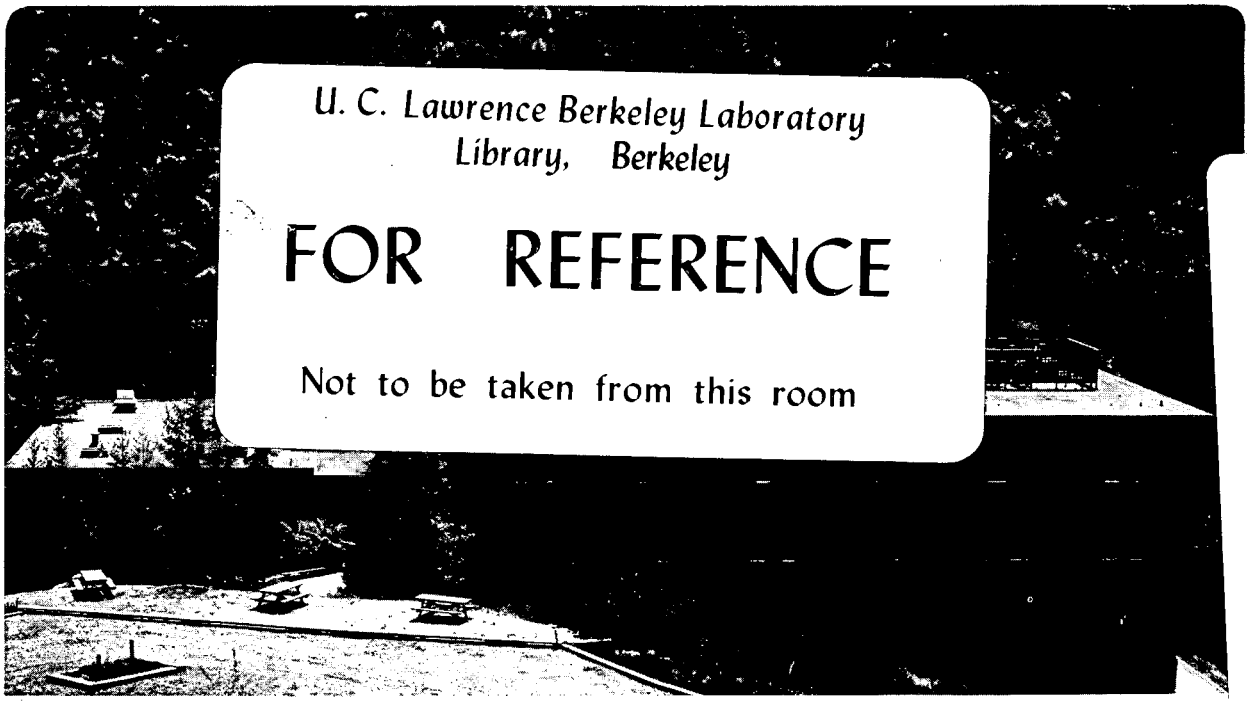
Materials Sciences Division

Presented at the Lattice Effects in High-T_c Superconductors Conference, Santa Fe, NM, January 13–15, 1992, and to be published in the Proceedings

The Influence of Anharmonic Phonons on the Isotope Effect in High-T_c Oxides

V.H. Crespi and M.L. Cohen

January 1992



U. C. Lawrence Berkeley Laboratory
Library, Berkeley

FOR REFERENCE

Not to be taken from this room

Copy 1
Bldg. 50 Library.

LBL-31865

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. Neither the United States Government nor any agency thereof, nor The Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or The Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or The Regents of the University of California and shall not be used for advertising or product endorsement purposes.

Lawrence Berkeley Laboratory is an equal opportunity employer.

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

LBL-31865

The Influence of Anharmonic Phonons on the Isotope Effect in High-T_c Oxides

Vincent H. Crespi and Marvin L. Cohen

Department of Physics
University of California

and

Materials Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

January 1992

This work was supported in part by the National Science Foundation under Grant No. DMR88-18404 and by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

The Influence of Anharmonic Phonons on the Isotope Effect in High-T_c Oxides

Vincent H. Crespi and Marvin L. Cohen

Department of Physics
University of California at Berkeley
Berkeley, California 94720
and
Materials Sciences Division
Lawrence Berkeley Laboratory
Berkeley, California 94720

Abstract

Anharmonic phonons are examined to study the unusual isotope effect exponents for the high-T_c oxides. Within a simple model of anharmonicity, the mass dependences of the electron-phonon coupling constant λ and the phonon frequency determine the isotope effect exponent α as a function of coupling strength. A model in which the outer wells of a multiple-well potential deepen as the orthorhombic/low temperature tetragonal phase transition in $La_{2-x}M_xCuO_4$ is approached is consistent with some experimentally observed variations in T_c and α.

As demonstrated by the other papers of this volume, there is ample evidence that anharmonic phonons are present in the high-T_c oxides. EXAFS data can be interpreted in terms of a double well for apical oxygen motion.[1] Ion channelling measurements indicate unusual atomic correlations below T_c.[2] The proximity of superconductivity to structural phase transitions,[3] especially in the $La_{2-x}M_xCuO_4$ systems, suggests the relevance of soft modes associated with these transitions. Pair distribution functions from neutron scattering data on $Nd_{2-x}Ce_xCuO_4$ and $Tl_2Ba_2CaCu_2O_8$ can be interpreted in terms of a superposition of two buckling distortions of O atoms perpendicular to the Cu-O bonds.[4] Frozen phonon calculations have yielded strongly anharmonic energy surfaces for tilts and rotations of the Cu-O octahedra in La_2CuO_4.[5] We will examine the possible effects of anharmonicity on the superconducting isotope effect in $La_{2-x}M_xCuO_4$ and $YBa_2Cu_3O_7$, with an eye towards accounting for the possibility of an isotope effect exponent greater than 0.5 in $La_{2-x}M_xCuO_4$.

The treatment of anharmonicity has been presented in previous papers.[6,7] In summary, an anharmonic expression for the electron-phonon coupling λ is constructed from a summation between the lattice ground state and all excited states for a dispersionless local oscillator at T=0[8].

$$\lambda = \sum_{n=1}^{\infty} N(0) \sum_{kk'}^{(FS)} \frac{|\langle n|M_{kk'}|0\rangle|^2}{E_n - E_0} \tag{1}$$

where $|k\rangle, |k'\rangle$ are electronic states on the Fermi surface and $|n\rangle$ is the phonon eigenstate of energy E_n. The electron-phonon matrix element $M_{kk'}$ is

$$M_{kk'} = \langle k'|V(r - \delta r) - V(r)|k\rangle, \tag{2}$$

where V(r) is the electron-ion potential. The electronic degrees of freedom are factorized out and lumped into a prefactor $\langle I^2\rangle$ which we will assume to be mass independent,

$$\lambda = N(0)\langle I^2\rangle \sum_{n=1}^{\infty} \frac{|\langle n|\delta r|0\rangle|^2}{E_n - E_0}. \tag{3}$$

Note that the electronic prefactor could obtain a mass dependence if the electronic structure is sensitive to fine details of structural distortions.[5] The summation is broken up into separate contributions to the electron-phonon coupling expression $\alpha^2 F(\omega)$

$$\alpha^2 F(\omega) = \sum_{n=1}^{\infty} \lambda_n \omega_n \delta(\omega - \omega_n), \tag{4}$$

where

$$\lambda_n = \langle I^2\rangle \left[\frac{|\langle n|\delta r|0\rangle|^2}{E_n - E_0}\right] \tag{5}$$

and

$$\omega_n = E_n - E_0. \tag{6}$$

This expression can be evaluated by numerical solution of the Schrödinger equation for the different isotopic masses. The isotope effect is then obtained from numerical solution of the Eliashberg equations. Note that the this treatment does not yield the value of λ, but only its mass dependence. The strength of the electron-phonon coupling is determined by the experimental results for T_c, at which point the isotope effect can be calculated.

The model is first applied to the higher temperature superconductors, for example $YBa_2Cu_3O_7$. As a simple model calculation, we consider a possible anharmonic potential modeling buckling motions of the planar oxygens perpendicular to the Cu-O bond. The potential has a quadrapolar form, with distance of Å between the wells and a well depth of 2000 K. Other potentials with differing widths or shallower wells produce similar results. Fitting to a value of $\lambda = 1.4$, we obtain a transition temperature of 110K and an isotope effect exponent of $\alpha = 0.04$. Large values of T_c and near-zero isotope effects are consistent with electron-phonon coupling within this model of anharmonic phonons.

The formalism can also be applied to the single-layer doped La_2CuO_4 compounds. As the strontium concentration increases in $La_{2-x}Sr_xCuO_4$, the isotope effect exponent appears to increase to values above 0.5 for $x \approx 0.12$ and then decrease to $\alpha \approx 0.1$ for greater strontium concentrations.[9] Meanwhile, T_c reaches a smooth maximum at $x \approx 0.15$. Barring questions of sample homogeniety, these variations can be analyzed in the context of potential anharmonic phonons associated with an incipient

low temperature orthorhombic to low temperature tetragonal structural phase transition. The approach to this transition is modelled by a quadrupolar potential with a central well and four outer wells associated with the four equivalent tilt directions for transition into the low teperature tetragonal phase. As the strontium concentration is increased, we assume that the outer wells deepen as shown in figure 1. For each

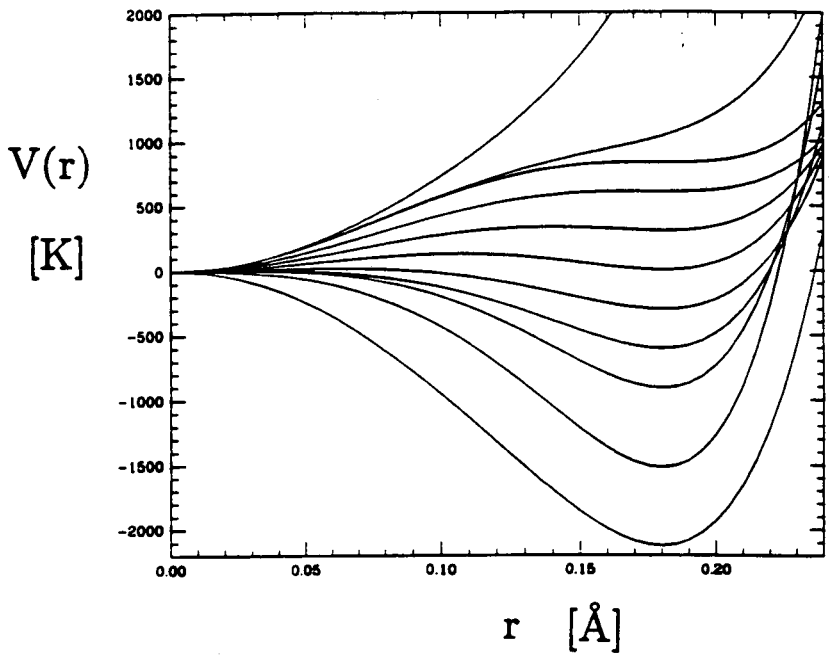


Figure 1: The r-dependence of the various quadrupolar potentials studied. Strontium concentration increases with increasing depth.

potential, the value of λ is adjusted to reproduce the experimental value of T_c. At this point, the model still contains limited freedom, in that the analysis does not provide a quantitative relation between strontium concentration and the particular potentials chosen; only the ordering of the potentials is fixed. After fitting to the experimental results for T_c, one obtains a variation in α with strontium concentration shown in figure 2. We emphasize that the agreement between theory and experiment is only qualitative due to the number of free parameters in the theory. A more complete discussion of these results is given in a previous publication.[7]

This choice for the evolution of the potentials assumes that the low temperature tetragonal phase becomes more favored with increasing strontium concentration. Qualitatively similar results could be obtained from a slightly different series of potentials which are identical to the potentials discussed for strontium concentrations less than x=0.12, but then flatten out for higher concentrations. This series of potentials produce a maximally stable low temperature tetragonal phase around x=0.12.

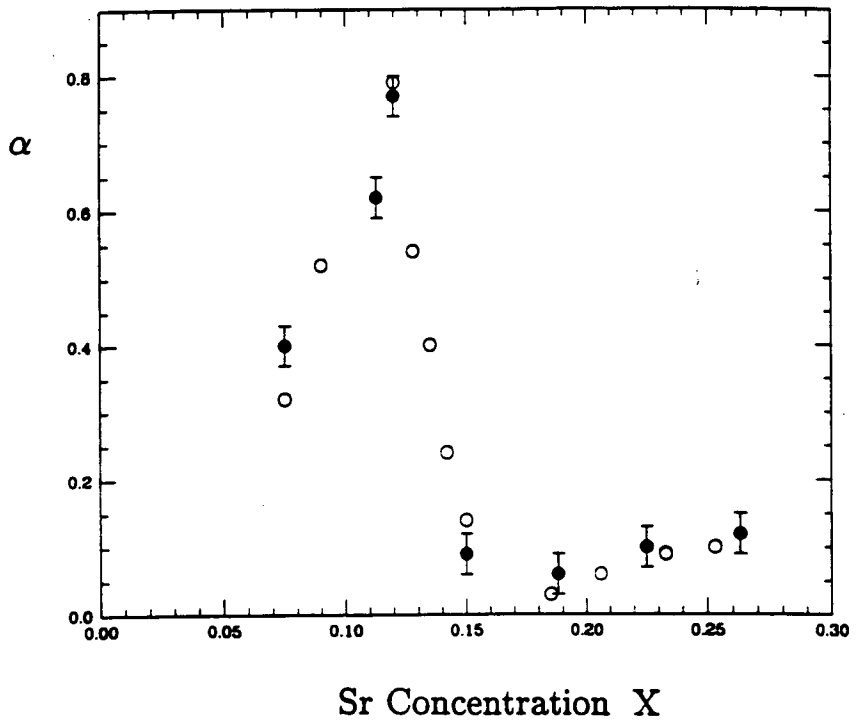


Figure 2: The isotope exponent α versus x in $La_{2-x}Sr_xCuO_4$ for the interionic potentials shown in Fig. 1. The solid circles are the data of Crawford et al.[9] The open circles are the theoretical results.

We note a potential ambiguity in the preceding discussion. The difficulty of defining normal modes in a strongly anharmonic system clouds the choice of effective oscillator mass. In the preceding analysis, the oscillator mass has been taken as the mass of an oxygen atom, instead of the reduced mass of the Cu-O octohedra, as would be valid for a harmonic normal mode. Such an assumption is plausible if the octahedra do not act as purely rigid entities. Assuming rigid motion of the Cu-O octahedra, the $La_{2-x}Sr_xCuO_4$ system can be reanalyzed within a more quantitative framework by solving the Schrödinger equation for the actual octopolar potential calculated from frozen phonon calculations[5] with an oscillator mass equal to the reduced mass of the Cu-O octahedra, namely 2.6 times the mass of an oxygen atom. In the case of purely quadratic electron-phonon coupling, a preliminary analysis yields an isotope effect exponent of $\alpha \approx 0.4$ for a rather large electron-phonon coupling of $\lambda \approx 3$. However, for such large values of the electron-phonon coupling the preceding zero-temperature treatment of anharmonicity is suspect, so that this result should be considered at best qualitative until a finite temperature treatment is performed. Recent experimental

work has suggested that the anomalous values of α in $La_{2-x}Sr_xCuO_4$ may be due to an isotope-dependent phase separation of a strontium-rich non-superconducting phase for copper-rich samples.[10] copper-poor samples, which do not show this phase separation, yield isotope effect exponents of roughly 0.4.

In summary, a simple model of anharmonicity can account for values of the isotope effect exponent both near zero and above 0.5 for superconducting transition temperatures in the range observed for the high-T_c oxides.

Acknowledgements

This research was supported by National Science Foundation Grant No. DMR88-18404 and by the Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U. S. Department of Energy under Contract No. DE-AC03-76SF00098. VHC was supported by a National Science Foundation Fellowship and a National Defense Science and Engineering Graduate Fellowship. CRAY computer time was provided by the Office of Energy Research of the U.S. Department of Energy at the Pittsburg supercomputing center.

References

1. J. Mustre de Leon, S. D. Conradson, I. Batistić and A. R. Bishop, *Phys. Rev. Lett.* **65** (1990) 1675 and the article by S. D. Conradson in this proceedings.

2. R. P. Sharma, L. E. Rehn, P. M. Baldo, and J. Z. Liu, *Phys. Rev. Lett.* **62** (1989) 2869 and the articles by L. E. Rehn and K. Yamaya in this proceedings

3. See the articles by J. D. Jorgensen, J. D. Axe, and M. K. Crawford in this proceedings.

4. See the article by S. J. L. Billinge in this proceedings.

5. W. E. Pickett, R. E. Cohen, H. Krakauer, *Phys. Rev. Lett.* **67** (1991) 228.

6. V. H. Crespi, M. L. Cohen and D. R. Penn, *Phys. Rev. B* **36** (1991) 12921.

7. V. H. Crespi and M. L. Cohen, *Phys. Rev. B* **44** (1991) 4712.

8. J. C. K. Hui and P. B. Allen, *J. Phys. F: Metal Phys.* **4** (1974) L42.

9. M. K. Crawford, M. N. Kunchur, W. E. Farneth, E. M. McCarron III, and S. J. Poon, *Phys. Rev. B* **41** (1990) 282.

10. M. Ronay, M. A. Frisch and T. R. McGuire, *Phys. Rev. B* **45** (1992) 335.

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
TECHNICAL INFORMATION DEPARTMENT
BERKELEY, CALIFORNIA 94720