Electron scattering from interacting tunneling units: A model for high-T_c superconductivity

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We consider the existence of high-T_c superconductivity and the symmetry of the gap function when electrons are scattered from tunneling units that interact via an elastic strainlike potential. We examine the consequences of conduction electron scattering for the specific case of tunneling units found in experiments on the high-T_c superconductors $YBa_2Cu_3O_{6+x}$, $Bi_2CaSr_2Cu_2O_8$, $Tl_2CaBa_2Cu_2O_8$, $Tl_2Ca_2Ba_2Cu_3O_{10}$, and $Tl_2CaBa_2CuO_6$. Our calculations give (i) a strongly anisotropic scattering of the conduction electrons, (ii) a strongly anisotropic superconducting gap in **k** space, (iii) an isotope effect different from that associated with phonon scattering in the BCS theory, (iv) a high transition temperature, and (v) a gap function with nodes and a combination of an s-wave and a $d_{x^2-y^2}$-wave symmetry. The $d_{x^2-y^2}$ symmetry arises from the directionally dependent scattering of electrons by the tunneling units which have a well-defined orientation with respect to the crystal axis. [S0163-1829(96)03033-0]

I. INTRODUCTION

There has been a great deal of interest in high-T_c superconductivity (SC) ever since it was discovered by Bednorz and Mueller.[1] The literature on high-T_c superconductivity is quite extensive[2–5] and therefore we only quote here a few review articles and books. Several theoretical explanations were proposed for high-T_c SC.[6–8] One of these ideas,[6] the resonance valence bond method, was used to explain high-T_c SC. Another[7–9] suggested that antiferromagnetic (AF) spin fluctuations are the cause of high-T_c SC. The theory of AF spin fluctuations gives an anisotropic gap function and nodes in the gap, which seems to have been confirmed in experiments.[10–12] Other models proposed were extended van Hove singularities,[13] anisotropic electron-phonon interactions,[14] and electron scattering from two-level tunneling units.[15,16]

The purpose of this paper is to propose an approach to the theory of high-T_c superconductivity.[17] We use the idea that the conduction electrons are scattered by atoms in a two-level (or multilevel) potential, denoted here as a tunneling unit (TU). The TU's interact with each other via a strainlike interaction. The orientation of the two-level TU, defined by the vector from one potential well to the other, has a well-defined direction with respect to the crystal axis. This idea is central to the understanding of our model.

For the high-T_c materials we consider, in Sec. II we review experimental evidence showing (a) the existence of tunneling units and (b) that the TU's are indeed aligned in a specific direction of orientation with respect to the crystal axis.

The idea that (noninteracting) double-well potentials may cause high-T_c superconductivity was discussed previously,[15,16] however, has not been widely accepted. For this reason it is important to elucidate the differences between our model and previous ones. Previous derivations[15,16] (i) did not consider interactions between tunneling units, (ii) relied on the concept that superconductivity is caused by anharmonic phonons arising when electrons are scattered from atoms in double-well potentials, (iii) did not give a d-wave-like symmetry for the gap function, which by now appears to be confirmed by experiments,[10–12] and (iv) gave an inverse (negative α) isotope effect.[18]

In our model electron pairing arises directly from scattering by the interacting tunneling units with no phonons involved; i.e., we assume that the phonon contribution is not the important one. Because of the experimentally observed direction of orientation of the TU's, we get an anisotropic scattering of the electrons and a combination of s-wave and d-wave symmetries in the gap function $\Delta(\mathbf{k})$. This s-wave and d-wave combination may be important in experiments on c-axis tunneling of pairs discussed in Sec. IX. Because of the interaction between the TU, we get asymmetric wells and obtain a usual isotope effect (positive α).

For the sake of clarity, we next discuss the physical ideas in more detail. Consider a single particle (i.e., an atom or a molecule) in a double-well potential at site i. Because the particle can tunnel from one well to the other, we denote this as a two-level tunneling unit TU(i) at site i. For simplicity we deal with a two-level TU; however, the principles derived, but not necessarily the details of the calculation, apply to multilevel TU's also. In fact we show later on that in a certain approximation the two-level and four-level TU's give the same symmetry of the gap function $\Delta(\mathbf{k})$. Tunneling units with elastic interactions were extensively discussed in connection with amorphous and glassy materials.[19–23] Here we explore the possibility of high-T_c superconductivity arising from electron scattering by tunneling units.[24]

The tunneling units are assumed to interact with other TU's as well as with nontunneling atoms or molecules in the high-T_c solid via a strainlike interaction.[19–23] The excitations arising from the long-range strainlike interactions[20–23] thus play an essential role in pairing and superconductivity. In Sec. VI we discuss that both the strain interaction and the single-electron–TU coupling are sufficiently strong to cause high-temperature superconductivity. The conduction electrons are scattered by the TU and excite it to a higher-

energy level, creating elastic excitations. A second electron deexcites the TU via a virtual process. This process can create an attractive electron-electron interaction, pairing, and SC. Furthermore, if the TU's in the high-T_c superconductor are oriented in a certain well-defined direction with respect to the crystal axis as is found in experiments on a number of high-T_c materials, the scattering results in an anisotropic electron–tunneling-unit potential. This anisotropic potential gives an anisotropic gap function with a combination of s and $d_{x^2-y^2}$ symmetry. Thus the anisotropy in the gap function comes out naturally from our calculations without any assumptions other than the existence of TU's which are oriented along a certain well-defined direction relative to the crystal axis.

TU's were found in experiments on a number of high-T_c materials. Experiments on $YBa_2Cu_3O_{6+x}$ showed that there are Cu-O chains along the b axis,[25,26,27] and that the oxygen atoms are displaced 0.08 Å perpendicular to the chain axis. For $YBa_2Cu_3O_{6+x}$ the direction of the TU's is in the a direction. TU's were also found in experiments on[26] $Bi_2Ca_1Sr_2Cu_2O_8$ where the oxygen atom occupies, and can tunnel between, one of the four possible sites, hence representing a four-level TU. Similarly[26] there are TU's in $Tl_2Ca_1Ba_2Cu_2O_8$, $Tl_2Ca_2Ba_2Cu_3O_{10}$, and $Tl_2Ca_0Ba_2Cu_1O_6$. Experiments thus show that TU's are present in a number of high-T_c materials. Are they present in *all* high-T_c materials? We cannot say at this time.

In Sec. VIII we discuss that in a certain approximation the two-level and four-level TU's give the same symmetry of the gap function $\Delta(\mathbf{k})$. Therefore we treat the case of $YBa_2Cu_3O_{6+x}$, which has two-level TU's on the chain, to obtain the symmetry of $\Delta(\mathbf{k})$.

II. TUNNELING-UNIT MODEL

We discuss the case of $YBa_2Cu_3O_{6+x}$ as a specific example of a high-T_c superconductor to which our derivations may apply. We assume the validity of the charge transfer model discussed by Jorgensen[28] and by Cava *et al.*[29,30] As was discussed by Jorgensen,[28] in $YBa_2Cu_3O_{6+x}$ the charge transfer layer has Cu-O chains along the b axis.[30] From neutron diffraction experiments Francois and co-workers[25–27] reported that in $YBa_2Cu_3O_{6+x}$ the oxygen atoms (in the charge transfer layer) on the Cu-O chains (along the b axis) are located in potential minima at a distance $d=0.08$ Å perpendicular to the chain axis (i.e., along the a axis) at temperatures from 5 to 250 K. Thus the oxygen atoms along the chains have two minimum positions perpendicular to the chain axis and are displaced from the chain axis by a well-defined distance in the a direction. A zigzag motion of the O(1) oxygens on the copper oxygen chains was also observed in Mössbauer experiments in the temperature region from 80 K to about[27] 400 K. The two positions seem to be occupied with equal probability at high temperatures. However, as the temperature is lowered the oxygen atoms move preferentially into one or the other of the two positions.[26] As stated in the Introduction, in a certain approximation the symmetry of $\Delta(\mathbf{k})$ for the chains in $YBa_2Cu_3O_{6+x}$ is the same as that for the TU's in the planes for $Tl_2Ca_1Ba_2Cu_2O_8$, $Tl_2Ca_2Ba_2Cu_3O_{10}$, and $Tl_2Ca_0Ba_2Cu_1O_6$.

Consider the left (right) potential wells for the oxygen. For simplicity we assume that with interactions the two potential wells have unequal depth. This idea is in qualitative agreement with the experiments that at high temperatures T the two potential minima are occupied equally likely; however, below some $T=T_0$ the oxygen atom prefers to be in one of the wells only.[25]

III. ZEROTH-ORDER HAMILTONIAN

To make a comparison between BCS theory[31] and our model, we write the zeroth-order Hamiltonian H_0 for both cases. Let $c_{\mathbf{k}}^*$ and $c_{\mathbf{k}}$ be the electron creation and annihilation operators (with the electron-spin index suppressed). Let $b_{\mathbf{q}}^*$ and $b_{\mathbf{q}}$ be the phonon creation and annihilation operators with wave vector $\mathbf{q}$. The zeroth-order Hamiltonian $H_0(\mathrm{BCS})$ for the BCS theory is $H_0(\mathrm{BCS}) = \Sigma_{\mathbf{k}} \epsilon_{\mathbf{k}} c_{\mathbf{k}}^* c_{\mathbf{k}} + \Sigma_{\mathbf{q}} \hbar \omega_{\mathbf{q}} b_{\mathbf{q}}^* b_{\mathbf{q}}$.

The zeroth-order TU Hamiltonian[19,20] $H_0(\mathrm{TU}) = H_{ce} + H_{\mathrm{st}}$, where H_{st} is the strain Hamiltonian, $H_{\mathrm{st}} = -\Sigma_{i<j} J_{ij} \sigma_i^z \sigma_j^z - \Sigma_i \xi_i \sigma_i^z$ where J_{ij} is the interaction potential between pairs of TU's at sites i and j, and σ_i^z is the z component of Pauli pseudospin operator. ξ_i is a local field at site i which acts as an external field.[20] ξ_i is related to the strain energy arising from nontunneling atoms and molecules.[20] Thus

$$H_0(TU) = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} c_{\mathbf{k}}^* c_{\mathbf{k}} - \sum_{i<j} J_{ij} \sigma_i^z \sigma_j^z - \sum_i \xi_i \sigma_i^z . \qquad (1)$$

In Eq. (1) the electron-spin index is again suppressed and only the pseudospin operators σ^z appear. Equation (1) for the tunneling units is the analog of $H_0(\mathrm{BCS})$ for the BCS model. We have not included the phonon contribution to the above Hamiltonian since we consider a physical situation in which the phonon excitations are much less important and negligible compared to the TU excitations. Instead of the phonon term in $H_0(\mathrm{BCS})$ we have the interaction term between the tunneling units, $-\Sigma_{i<j} J_{ij} \sigma_i^z \sigma_j^z - \Sigma_i \xi_i \sigma_i^z$. Thus instead of the phonon energies in the BCS theory, the TU energies enter in our derivations.

IV. INTERACTION WITH THE CONDUCTION ELECTRONS

In BCS theory, the electron-phonon interaction Hamiltonian $H_{\mathrm{int}}(ep)$ between the conduction electrons and the phonons is given by $H_{\mathrm{int}}(ep) = \Sigma_{\mathbf{k},\mathbf{q}} V_{\mathbf{q}} c_{\mathbf{k}}^* c_{\mathbf{k}+\mathbf{q}} (b_{\mathbf{q}}^* + b_{-\mathbf{q}})$. We next obtain the conduction electron TU interaction Hamiltonian $H_{\mathrm{el,TU}}$, which is analogous to $H_{\mathrm{int}}(ep)$ for the BCS case.

Let $\mathbf{r}$ and $\mathbf{R}_i$ be the position of the electron and of the TU site i, respectively. Let the $V_0(\mathbf{r}-\mathbf{R}_i)$ be the Coulomb potential between the TU and the electron. The oxygen atom experiences a double-well potential and can jump (or tunnel) between the two wells. The conduction electron which scatters the oxygen atom feels a potential which has a certain magnitude but whose center varies as a result of the jump of the atom. When the atom jumps from the left well into the right one or vice versa, the wave function of the conduction electron follows it. A similar problem was treated by

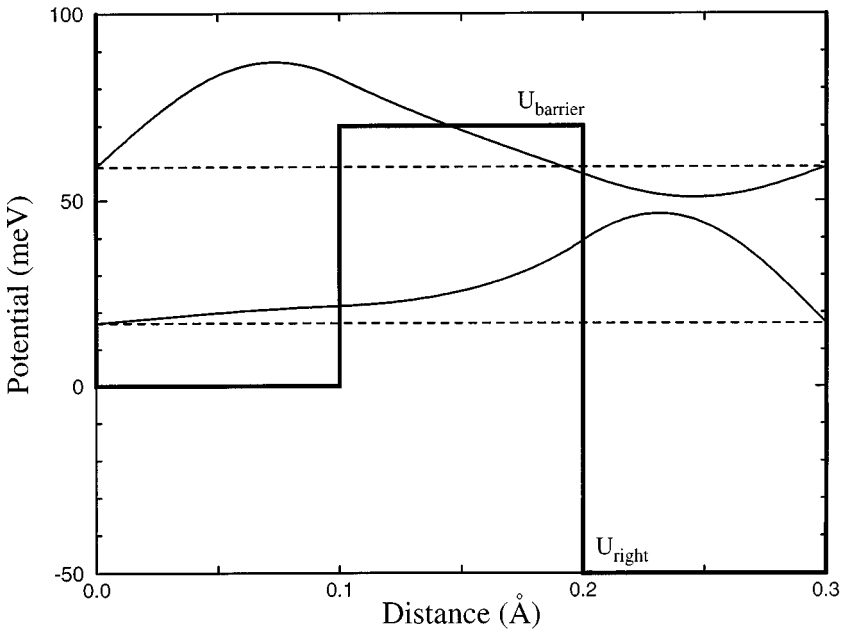

FIG. 1. A model potential for calculating the overlap integral given by Eq. (7). The figure also shows the wave functions for the two lowest energies of the tunneling unit. U_{barrier} and U_{right} are measured from the bottom of the left well, taken to be zero.

Kondo[32] and we follow Kondo's approach.

The electron wave function is assumed to be of the free electron type $\exp(i\mathbf{k}\cdot\mathbf{r})$. $V_0(\mathbf{r}-\mathbf{R}_i)$ is assumed to depend only on the relative coordinates $(\mathbf{r}-\mathbf{R}_i)$. The oxygen atom has two localized states located at sites $+a/2$ or $-a/2$ from the center. The distance $a/2$ is a vector distance chosen to be along the x direction, where $a/2$ is measured from the chain axis.

Let $\Psi_R(\mathbf{R})$ and $\Psi_L(\mathbf{R})$ be the wave functions when the particle is in the ground and excited states, respectively. These states are localized preferentially in the right (left) wells of the double-well potential. For clarity the index i is suppressed. Then we have[32]

$$H_{\text{el,TU}}=\sum_{\mathbf{k}'\mathbf{k}}(L_{\mathbf{k}'\mathbf{k}}+M_{\mathbf{k}'\mathbf{k}}\sigma^z+N_{\mathbf{k}'\mathbf{k}}\sigma^x)c^*_{\mathbf{k}'}c_{\mathbf{k}}, \tag{2}$$

where σ is the Pauli pseudospin matrix expressed in the basis functions Ψ_L and Ψ_R ($\sigma_x^2=\sigma_y^2=\sigma_z^2=1$) and[32]

$$L_{\mathbf{k}'\mathbf{k}}=V_0(\mathbf{k}'-\mathbf{k})\int e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}}\frac{\Psi_L(\mathbf{R})^2+\Psi_R(\mathbf{R})^2}{2}d^3\mathbf{R}, \tag{3}$$

$$M_{\mathbf{k}'\mathbf{k}}=V_0(\mathbf{k}'-\mathbf{k})\int e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}}\frac{\Psi_R(\mathbf{R})^2-\Psi_L(\mathbf{R})^2}{2}d^3\mathbf{R}, \tag{4}$$

$$N_{\mathbf{k}'\mathbf{k}}=V_0(\mathbf{k}'-\mathbf{k})\times\int e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}}\frac{\Psi_L^*(\mathbf{R})\Psi_R(\mathbf{R})+\Psi_R^*(\mathbf{R})\Psi_L(\mathbf{R})}{2}d^3\mathbf{R}. \tag{5}$$

$H_{\text{el,TU}}$ is the Hamiltonian for electron scattering by a single TU. The momentum due to the scattering is taken up by the excitation of the system of interacting TU's and will be discussed later on. The $L_{\mathbf{k}'\mathbf{k}}$ (renormalization of the Fermi energy) and $M_{\mathbf{k}'\mathbf{k}}$ (renormalization of the TU energy) terms do not produce virtual excitations of the TU and therefore are not relevant to the pairing of electrons and superconductivity. The third term in Eq. (2) represents the physical process in which an incoming electron with wave vector $\mathbf{k}$ is annihilated (scattered) by a TU which has an electron–tunneling-unit interaction potential $V_0(\mathbf{k}'-\mathbf{k})$. Another electron is created with wave vector $\mathbf{k}'=\mathbf{k}+\mathbf{q}$. In this process the particle in one state of the TU is flipped to the other state as is indicated by the σ^x operator. Thus, the $N_{\mathbf{k}'\mathbf{k}}$ term represents the electron scattering followed by an excitation of the tunneling unit from the ground state to the excited state at site i. The total Hamiltonian is given by summing over all tunneling sites i in the solid. The effective Hamiltonian $H_{\text{int}}(\text{el,TU})$ is

$$H_{\text{int}}(\text{el,TU})=\sum_{\mathbf{k}'\mathbf{k},i}N_{\mathbf{k}'\mathbf{k},i}\sigma_i^x c^*_{\mathbf{k}'}c_{\mathbf{k}}, \tag{6}$$

where

$$N_{\mathbf{k}'\mathbf{k},i}=V_0(\mathbf{k}'-\mathbf{k})\int e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i}[\Psi_L^*(\mathbf{R}_i)\Psi_R(\mathbf{R}_i)+\text{H.c.}]\frac{d^3\mathbf{R}_i}{2}. \tag{7}$$

Equation (6) is the electron-TU interaction term, analogous to the electron-phonon term in for the BCS superconductor and H.c. denotes the Hermitian conjugate.

V. ANISOTROPY IN THE SCATTERING POTENTIAL

We now show that for the $YBa_2Cu_3O_{6+x}$ chains Eq. (7) gives an anisotropic potential which results in an anisotropic gap function $\Delta(\mathbf{k})$ with a combination of s-wave and d-wave symmetries.[33,34] Each TU is directed in the a direction, denoted as the x direction. The major contribution to

tunneling occurs for large overlap integrals, i.e., between the two minimum positions for the oxygen on the same site.[33] This is so, since the overlap integral between wells on the opposite side of the copper atom in the chain is very small due to the large distance between wells. We approximate the overlap in Eq. (7) by the delta functions $\delta(y)$ and $\delta(z)$ in the y and z directions. Thus the scattering potential is strongly directional.

To simulate the experimental results[25] for the oxygen tunneling between two positions located perpendicular to the copper-oxygen chain we use the model potential shown in Fig. 1. The potential of the left well was assumed to be 50 meV above that of the right well and the height of the barrier was assumed to be 70 meV. We calculated[35] the wave functions $\Psi_L(\mathbf{R}_i)$ and $\Psi_R(\mathbf{R}_i)$ and plotted them in Fig. 1. The width of the two wells and the width of the barrier were each assumed to be 0.1 Å. For this potential the ground-state wave function is mostly localized in the right well; in the next lowest state the oxygen is mostly in the left well. The next excited state is no longer localized (its energy is above the barrier). The two lowest energies corresponding to the two wave functions shown in Fig. 1 are 17 meV and 59 meV. The overlap integral $N_{\mathbf{kk}'}/V_0(\mathbf{k}-\mathbf{k}')$ was calculated and plotted as a function of the angles, θ and θ' between the x axis and the wave vectors $\mathbf{k}$ and $\mathbf{k}'$. Our calculations give that the overlap integral is proportional to $\cos\theta-\cos\theta'$, showing that

$$N_{\mathbf{kk}'} \propto V_0(\mathbf{k}-\mathbf{k}')(\cos\theta-\cos\theta')e^{-\sqrt{2mU}\Delta x/\hbar}, \tag{8}$$

where U in Eq. (8) is some average potential barrier and Δx is the distance between the wells. Assuming the isotropic Coulomb interaction $V_0(\mathbf{k}-\mathbf{k}')$ we have

$$N_{\mathbf{kk}'}=N_i(\cos\theta-\cos\theta'), \tag{9}$$

where N_i is an isotropic coefficient which incorporates the Gamow factor from Eq. (8). Equation (9) will be used to solve the tunneling problem.

It is important to note that the interaction of a pair of two-level TU's via the conduction electron scattering was discussed in a number of articles related to the Kondo effect.[36,37] Let this interaction potential be denoted by $V_{(\mathrm{TU,TU})}$. $V_{(\mathrm{TU,TU})}$ is somewhat similar to the interaction potential arising between localized magnetic impurities in the presence of an $\mathbf{s}\cdot\mathbf{d}$ scattering of the conduction electrons by the localized magnetic impurities. This $\mathbf{s}\cdot\mathbf{d}$ scattering leads to the well-known Ruderman-Kittel[38] potential between the localized spins.

It was found that the interaction between pairs of two-level TU's, $V_{(\mathrm{TU,TU})}$, arising purely from conduction electron scattering is quite weak.[37] It is important to understand that the indirect interaction between TU's, $V_{(\mathrm{TU,TU})}$, arising from conduction electron scattering is not relevant to our problem, and thus it does not matter that $V_{(\mathrm{TU,TU})}$ is weak, since we assume that the interaction potential between the TU's arises from the strain interaction. We discuss a *strongly interacting* system[39] in which the coupling due to strain interaction is much greater than $V_{(\mathrm{TU,TU})}$. Hence in our problem $V_{(\mathrm{TU,TU})}$ can be neglected compared to the strain interaction.

VI. EXCITATION ENERGY FOR THE TUNNELING UNITS

In weak-coupling BCS theory one obtains an attractive electron-electron interaction and the transition temperature using second-order perturbation theory, whereas in the strong-coupling case one has to use the Eliashberg[40] equations. We treat the electron-TU scattering problem in the weak-coupling approximation.[41]

In the BCS Hamiltonian the electron-phonon scattering results in superconducting pairing. The excitation energy $\hbar\omega_q$ is found from the equation of motion of the electron-phonon interaction term with H_0(BCS)

To obtain the effective interaction between pairs of electrons scattered by TU's we have to derive the excitation spectrum of the interacting TU system. What range of excitation energies are we interested in? From treating the interacting random quadrupole system[20–22] it was found that for the random amorphous system there are very-low-energy excitations of the order of 1 K and other excitations which arise from the frozen-in system of TU's that have much higher energies. The largest energy is determined by the total strain energy of the TU with its surrounding. In the high-T_c superconductor we are not dealing with a random system and we are concerned here primarily with the high-energy excitation of the system because these excitations will give the major contribution to the attractive interaction and the high transition temperature (there may still be some low-energy excitations).

The algebra here is very similar to the one in the case of the phonons.[42] $N_{\mathbf{kk}'}$ replaces the $D(q)$ for the phonons, and $\hbar\omega_q$ for the phonons is replaced by the TU excitation energy $J_s=(2\Sigma_i J_{si}-\xi_s)$ derived in the Appendix. In our calculation we use J_{ave}, the average value of J_s. We obtain the expression[42] for the electron-electron interaction Hamiltonian $H_{\mathrm{el,el}}$,

$$H_{\mathrm{el,el}}=\sum_{k,q} N(q)N(-q) \times \frac{J_{\mathrm{ave}}}{(\epsilon_k-\epsilon_{k+q})^2-J_{\mathrm{ave}}^2} c^*_{k+q}c^*_{-k-q}c_{-k}c_k . \tag{10}$$

We find that for $(\epsilon_k-\epsilon_{k+q})^2<J_{\mathrm{ave}}^2$ $H_{\mathrm{el,el}}$ is attractive, and causes pairing. The interacting tunneling system takes up the momentum when the particle is scattered from one well to the other. When there are no phonons excited, all the momentum is taken up by the interacting tunneling system. In principle, however, part of the momentum could be transferred to phonon excitations, in which case the interacting system will take up only *part* of the momentum imparted to the TU.

It is important to estimate the strength of the average strain-interaction potential J_{ave}. Whereas calculations by Brown[39] show the presence of relatively large strains, we could not estimate the strain energy for the high-T_c superconductors from Brown's paper[39] directly. However, experiments on the insulating strain-interacting TU's of CN^- dissolved in KBr (Refs. 43 and 44) show that estimates of the

near-neighbor strain interaction J_{NN} are large and vary somewhat in different experiments. Experiments by Enss *et al.*[43] obtain the strength of the near-neighbor strain interaction J_{NN} from the low-energy excitations of CN^- in KBr, and find that J_{NN} is about 300 K.[44] Dobbs *et al.*[45] obtain J_{NN} from low-temperature specific heat experiments and find that J_{NN} may be as high as 1000 K.[45] Assuming J_{NN} is of similar magnitude in the high-T_c materials as in CN^- in KBr, we obtain that if strains from two or more near neighbors act on the TU under consideration, the high-energy excitation is at least 600 K but may be even much higher than that. The single-electron–tunneling-unit scattering potential which enters our equations can also be quite large. For one particular case, involving the glassy metal $Pd_{0.75}Si_{0.165}Cu_{0.06}$,[46] this potential was deduced from experiments on electron relaxation in metallic glasses and was found to be about 0.4 eV. Using the value for J_{ave} of 600 K and the value of $N(q)$ to be 0.4 eV we obtain a coupling constant which gives a high T_c. We also have to consider the number of TU's which are present in the crystal. If the TU concentration were very low, as it is in glasses, we would most likely not get high-T_c superconductivity. However, here we expect the concentration of the order of one tunneling unit per unit cell, thus enabling the possibility for high T_c. There may be additional low-energy excitations[20] in the strain interacting system; however, here T_c will be determined by the high-energy excitations present in the system. Our estimates indeed give that the strain interactions may be large enough to cause high-T_c superconductivity.

VII. EQUATIONS FOR THE GAP FUNCTION AND T_c

We next derive the expression for the gap function $\Delta(\mathbf{k})$. We start with an effective electron-electron potential, denoted by $V(\theta,\theta')$. $V(\theta,\theta')$ consists of two parts, a repulsive interaction V_1 arising from the Coulomb interaction between pairs of electrons as they are scattered by the TU's, without the TU's being excited, and a second part in which the TU is virtually scattered from one state to the other, causing an attractive electron-electron interaction given by Eq. (10). Substituting Eq. (8) into Eq. (10) we have

$$V(\theta,\theta')=V_1+V_2(\cos\theta-\cos\theta')^2. \tag{11}$$

We assume that the gap function can be written as

$$\Delta(\theta)=\Delta_1+\Delta_2\cos^2\theta. \tag{12}$$

We use the standard expression $\Delta(\mathbf{k})=\Sigma_{\mathbf{k}'}V_{\mathbf{k}\mathbf{k}'}\Delta(\mathbf{k}')$ for the finite-temperature integral equation for the gap function[31] and obtain

$$\Delta_1+\Delta_2\cos^2\theta=\frac{N_0}{4\pi}\int_{-J}^{J}d\epsilon'\int_{-\pi}^{\pi}d\theta'\,\frac{(V_1+V_2\cos^2\theta'+V_2\cos^2\theta)(\Delta_1+\Delta_2\cos^2\theta')}{\sqrt{\epsilon'^2+(\Delta_1+\Delta_2\cos^2\theta')^2}}\tanh\frac{\beta\sqrt{\epsilon'^2+(\Delta_1+\Delta_2\cos^2\theta')^2}}{2}, \tag{13}$$

where J is the maximum TU excitation energy, similar to the cutoff energy $\hbar\omega_c$ in BCS calculations, and N_0 is the density of states at the Fermi surface. Equation (13) can be separated into a system of two equations for Δ_1 and Δ_2:

$$\Delta_1=\frac{N_0}{4\pi}\int_{-J}^{J}d\epsilon'\int_{-\pi}^{\pi}d\theta'\,\frac{(V_1+V_2\cos^2\theta')(\Delta_1+\Delta_2\cos^2\theta')}{\sqrt{\epsilon'^2+(\Delta_1+\Delta_2\cos^2\theta')^2}}\tanh\frac{\beta\sqrt{\epsilon'^2+(\Delta_1+\Delta_2\cos^2\theta')^2}}{2}, \tag{14}$$

$$\Delta_2=\frac{N_0}{4\pi}\int_{-J}^{J}d\epsilon'\int_{-\pi}^{\pi}d\theta'\,\frac{V_2(\Delta_1+\Delta_2\cos^2\theta')}{\sqrt{\epsilon'^2+(\Delta_1+\Delta_2\cos^2\theta')^2}}\tanh\frac{\beta\sqrt{\epsilon'^2+(\Delta_1+\Delta_2\cos^2\theta')^2}}{2}. \tag{15}$$

At $T=T_c$ both $\Delta_1=0$ and $\Delta_2=0$. Let b be the limit of Δ_2/Δ_1 as $T\to T_c$. Dividing Eqs. (14) and (15) by Δ_1 we obtain the following pair of equations for b and T_c:

$$1=\frac{N_0}{2\pi}\int_0^J d\epsilon'\int_{-\pi}^{\pi}d\theta'\times\frac{(V_1+V_2\cos^2\theta')(1+b\cos^2\theta')}{\epsilon'}\tanh\frac{\beta_c\epsilon'}{2}, \tag{16}$$

$$b=\frac{N_0}{2\pi}\int_0^J d\epsilon'\int_{-\pi}^{\pi}d\theta'\,\frac{V_2(1+b\cos^2\theta')}{\epsilon'}\tanh\frac{\beta_c\epsilon'}{2}. \tag{17}$$

Integrating over θ' yields

$$1=N_0\left(V_1+\frac{V_2+bV_1}{2}+\frac{3}{8}bV_2\right)\int_0^J\frac{d\epsilon}{\epsilon}\tanh\frac{\beta_c\epsilon}{2}, \tag{18}$$

$$b=N_0V_2\left(1+\frac{b}{2}\right)\int_0^J\frac{d\epsilon}{\epsilon}\tanh\frac{\beta_c\epsilon}{2}. \tag{19}$$

Dividing Eq. (19) by Eq. (18) we obtain

$$b=\frac{V_2(1+0.5b)}{V_1(1+0.5b)+0.5V_2(1+0.75b)},$$

$$0=\left(\frac{1}{2}V_1+\frac{3}{8}V_2\right)b^2+V_1b-V_2,$$

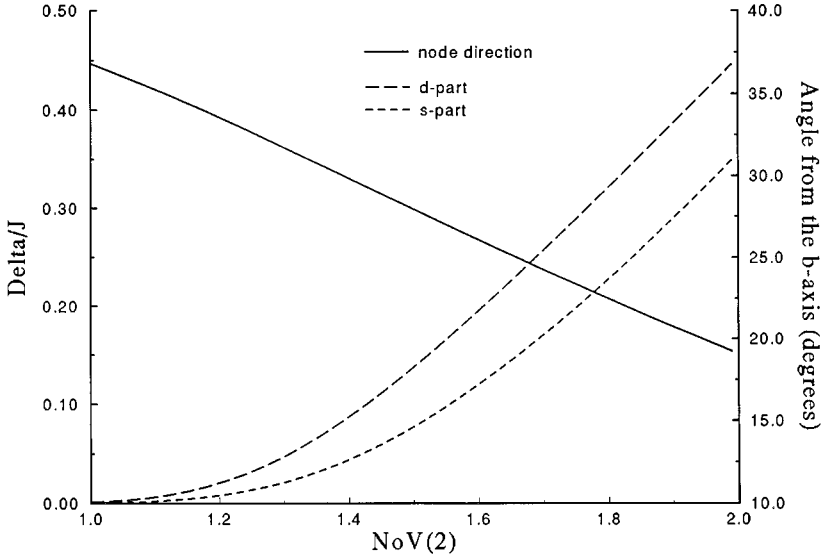

FIG. 2. Gap function coefficients Δ_s and Δ_d (measured in units of J) as functions of N_0V_2 for a fixed value of $N_0V_1=-1.8$. The solid line shows the direction of nodes in the gap function $[\Delta(\mathbf{k})=0]$ with respect to the b axis.

$$b=\frac{-V_1\pm\sqrt{(V_1+V_2)^2+0.5V_2^2}}{V_1+0.75V_2}. \tag{20}$$

Substituting Eq. (20) into Eq. (19) and dividing it by b gives us the final BCS-like equation for T_c:

$$1=N_0V_{\text{eff}}\int_0^J\frac{d\epsilon}{\epsilon}\tanh\frac{\beta_c\epsilon}{2}. \tag{21}$$

We thus have

$$k_BT_c\approx Je^{-1/(N_0V_{\text{eff}})}, \tag{22}$$

where

$$V_{\text{eff}}\equiv V_2\left(\frac{1}{b}+\frac{1}{2}\right)=V_2\left(\frac{1}{2}+\frac{V_1+0.75V_2}{-V_1+\sqrt{(V_1+V_2)^2+0.5V_2^2}}\right). \tag{23}$$

We chose the plus sign in Eq. (20) because it yields a higher effective potential V_{eff} for any combination of V_1 and V_2. In Eq. (23), V_1 is negative (repulsive interaction). Equation (12) can be rewritten to show the explicit dependence of the gap function upon $\cos2\theta$. We have

$$\Delta(\theta)=\Delta_s+\Delta_d\cos2\theta, \tag{24}$$

where $\Delta_s=\Delta_1+\Delta_2/2$ and $\Delta_d=\Delta_2/2$. The coefficient Δ_d of $\cos2\theta$ gives the amplitude of the $d_{x^2-y^2}$ part of the gap function. The gap function $\Delta(\theta)$ has nodes when Δ_d is greater than Δ_s.

VIII. SOLUTION FOR THE ZERO-TEMPERATURE GAP EQUATION

For zero temperature we solved the integral equations (14) and (15) and constructed algebraic equations which closely approximate the computer calculated results. The coefficients Δ_s and Δ_d for the zero-temperature gap function were obtained as functions of the parameters N_0V_1 and N_0V_2. Figure 2 shows Δ_s and Δ_d vs N_0V_2 for a fixed value

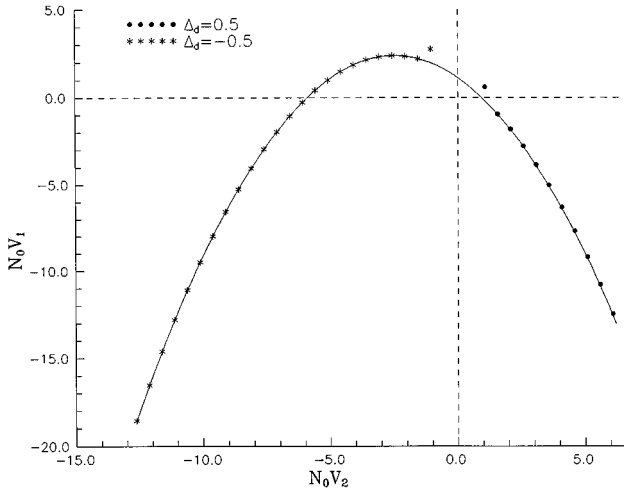

FIG. 3. Line of constant value $|\Delta_d/J|=0.5$. The solid line shows the best-fit parabola.

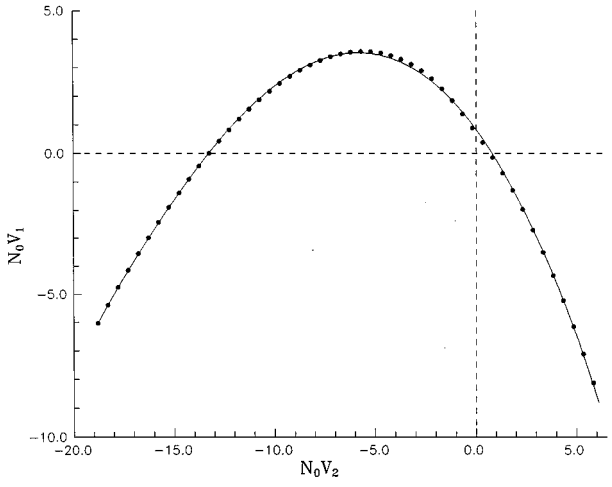

FIG. 4. Line of constant value $\Delta_s/J=0.5$. The solid line shows the best-fit third-degree polynomial.

of $N_0V_1=-1.8$. It also shows direction of the nodes in the gap function $\Delta(\mathbf{k})$.

While our model deals with repulsive (negative) N_0V_1 and attractive (positive) N_0V_2, we used a wide range of both negative and positive potentials N_0V_1 and N_0V_2 to examine the mathematical properties of Eqs. (14) and (15). We plotted isolines of Δ_s and Δ_d (lines of constant Δ_s and of constant Δ_d) using N_0V_1 and N_0V_2 as coordinates. For clarity we denote $u_1=N_0V_1$ and $u_2=N_0V_2$. Some of these plots are shown in Figs. 3 and 4.

For $0.1J\leqslant\Delta_s\leqslant0.5J$ the lines of constant Δ_s can be approximated by third-degree polynomials in u_1 and u_2 with very high accuracy as is shown in Fig. 4. Coefficients of these polynomials can in turn be analyzed as functions of Δ_s, leading to the approximate algebraic equation

$$u_1\approx1.57\Delta_s+(1-0.032\ln\Delta_s)u_2+(0.0414+0.0716\ln\Delta_s)u_2^2+(-0.000\,114+0.001\,49\ln\Delta_s)u_2^3. \tag{25}$$

The lines of constant Δ_d have somewhat different behavior. In the vicinity of $u_2=0$ these isolines go almost parallel to the N_0V_1 axis. The reason for this is that for a system with spherical symmetry ($N_0V_2=0$) $\Delta_d=0$. For an assumed repulsive anisotropic potential [this is a mathematical assumption not within our TU model which always gives an attractive (positive) anisotropic potential] one can derive the important feature that Δ_d has always the same sign as N_0V_2. The lobes of the d-wave component of the gap function change sign if one uses an (assumed) repulsive anisotropic potential instead of an attractive one. When the scattering is preferred to be in the x direction and the potential N_0V_2 is attractive the d-wave lobes are positive in the x direction and negative in the y direction, whereas for a repulsive anisotropic potential N_0V_2 with the same preferred direction of scattering (x direction), the lobes in the x direction are negative and those in the y direction are positive. To simplify the picture let us concentrate on a sufficiently large $|u_2|>u_{2\min}$, where $u_{2\,\min}$ varies from the highest value of 0.8 for $|\Delta_d|=0.5J$ to lower values for smaller d-wave components. Then for any $|\Delta_d|$ *both* positive and negative branches of the remaining $|\Delta_d|$ isoline can be very accurately approximated by *the same* second-degree polynomial in u_1 and u_2. Analyzing coefficients of these polynomials as functions of $|\Delta_d|$ leads to the algebraic expression

$$u_1\approx(0.251+1.88|\Delta_d|)-u_2+(-0.118+0.121\ln|\Delta_d|)u_2^2. \tag{26}$$

Equations (25) and (26) can be rewritten in a form which can be easily solved for Δ_d and Δ_s,

$$|\Delta_d|\approx e^{8.39(u_1+u_2-0.251+0.118u_2^2-1.88|\Delta_d|)/u_2^2}, \tag{27}$$

$$\Delta_s\approx e^{13.9(u_1+u_2-0.0414u_2^2-1.57\Delta_s)/(-0.446u_2+u_2^2+0.0207u_2^3)}. \tag{28}$$

Now we return to discuss the relation between the two-orientational tunneling units and the four-orientational ones and show that in a certain approximation the two give the same symmetry for the gap function.

Consider a four-well TU with well coordinates $\mathbf{r}_1=(-a,0)$, $\mathbf{r}_2=(0,a)$, $\mathbf{r}_3=(0,-a)$, and $\mathbf{r}_4=(a,0)$. We assume that in the ground state the oxygen atom is in the first well at $\mathbf{r}_1$. We also assume that the wells located at $\mathbf{r}_2$ and $\mathbf{r}_3$ have the same potential, that this potential is higher than that of the first well, and that well number 4 has a potential different from wells 1, 2, and 3. If $\Psi_i(\mathbf{R})$ is the wave function of the oxygen localized in the ith well, then the ground-state wave function is $\Psi_g(\mathbf{R})=\Psi_1(\mathbf{R})$ and possible excited-state wave functions are $[\Psi_2(\mathbf{R})+\Psi_3(\mathbf{R})]/\sqrt{2}$, $[\Psi_2(\mathbf{R})-\Psi_3(\mathbf{R})]/\sqrt{2}$, and $\Psi_4(\mathbf{R})$. Coupling to $\Psi_4(\mathbf{R})$ is small due to the large exponent in the Gamow factor. From the two remaining states the symmetric one always has lower energy; therefore we must use $\Psi_e(\mathbf{R})=[\Psi_2(\mathbf{R})+\Psi_3(\mathbf{R})]/\sqrt{2}$ as the wave function for the excited state in the coupling integral similar to Eq. (2). Let $N_{\mathbf{q},ij}$ be the function $N_{\mathbf{q}}$ when the oxygen is scattered from state i to state j. Using $\mathbf{q}\equiv\mathbf{k}-\mathbf{k}'$ the resulting coupling constant can then be expressed as $N_{\mathbf{q}}=(N_{\mathbf{q},12}+N_{\mathbf{q},13})/\sqrt{2}=(1/\sqrt{2})\times[N_{\rm isot}(q_x+q_y)/\sqrt{2}+N_{\rm isot}(q_x-q_y)/\sqrt{2}]=N_{\rm isot}q_x$, where we have used the fact that $N_{\mathbf{q},ij}\propto N_{\rm isot}[\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)]$.

For the above argument to be valid, the lowest-energy state of the TU should not change direction from one site to the other, for if it did change direction, the angle θ would vary from site to site. Consider the case of $Bi_2Ca_1Sr_2Cu_2O_8$. For this material there are four possible wells in which the oxygen can be found. The microscopic symmetry, as determined by electron beam diffraction, is orthorhombic.[26] Thus the lattice distorts, removing the tetragonal symmetry of the four orientations and introducing a preferred orientation for the ground state of the oxygen. An orthorhombicity of the order of a small percent is sufficient to have preferred directions of orientation, since the characteristic length is the distance between wells on the same site. This results in a factor of q_x in the expression for $N_{\mathbf{q},ij}$.

IX. POSSIBLE EXPERIMENTAL CONSIDERATIONS

Is it possible to obtain experimental verification of our calculations? Further theoretical as well as experimental work needs to be done to decide this question. There are, however, two possible experiments that could shed light on our calculations. These are (i) the pressure dependence of the transition temperature and (ii) the isotope effect.

If the strain interactions are important to high-T_c superconductors, one should be able to observe the change in T_c as the strain interaction is varied, assuming we keep the temperature of the material very close to T_c and apply a slowly varying-time dependent pressure. Since the near-neighbor strain interactions, which primarily determine the transition temperature, are proportional to r^{-3}, as the pressure is changed, so is the distance between the atoms, and one could drive the material from superconducting to normal and vice versa as the pressure is slowly varied. This is one possible experiment which may be very relevant to our results.

A. Calculation of the isotope effect

Another possible experiment is the isotope effect. The isotope effect arising from a particle in a double-well potential has been discussed previously.[18] In these earlier calcula-

tions the isotope effect was derived from the mass-dependent coupling of electrons to excitations of an atom in a symmetric double-well potential described in terms of anharmonic phonons. In a similar fashion, we use the electron-TU coupling term expressed in Eq. (7) to calculate the isotope effect for our model. The major difference is that we consider asymmetric potential wells due to the interaction between the tunneling units. This leads to very different results.[47]

Here we present a brief derivation of the isotope effect $\alpha = -\partial \ln T_c / \partial \ln m$ for our model. Starting from Eq. (22) we have $\ln T_c = \ln(J/k_B) - (N_0 V_{\rm eff})^{-1}$. As seen in Eq. (23) the effective potential $V_{\rm eff}$ depends on two terms V_1 and V_2. We assume that there is no isotope effect associated with electron-electron Coulomb interaction V_1 and the strain interaction J. Thus we have

$$\alpha = -\frac{m}{N_0 V_{\rm eff}^2}\frac{\partial V_{\rm eff}}{\partial V_2}\frac{\partial V_2}{\partial m}. \tag{29}$$

Because of the first partial derivative term, the isotope effect partly depends on the relative importance of the two terms V_1 and V_2 in the expression for $V_{\rm eff}$ in Eq. (23). V_2 arises from the effective electron-electron Hamiltonian, Eq. (10). The mass dependence of V_2 comes from $|N(q)|^2$ where the electron-TU coupling term $N(q)$ is given by Eq. (7). This term depends on the mass of the particle through the wave functions Ψ_L and Ψ_R. The final expression for the isotope effect can therefore be written as

$$\alpha = -\frac{m N_0 V_2}{(N_0 V_{\rm eff})^2}\left(\frac{V_{\rm eff}}{V_2} - \frac{1.5\sqrt{(V_1+V_2)^2+0.5V_2^2}+0.5(V_1+V_2)}{2\sqrt{(V_1+V_2)^2+0.5V_2^2}\,[V_1-\sqrt{(V_1+V_2)^2+0.5V_2^2}]^2}V_1V_2\right)\frac{\partial \ln |N(q)|^2}{\partial m}. \tag{30}$$

We have evaluated the isotope effect α for a model potential shown in Fig. 1 varying both the depth of the well, $U_{\rm right}$, and the barrier height $U_{\rm barrier}$. We solve for the exact eigenfunction for the oxygen atom in the double-well potential for a continuously variable oxygen mass to numerically calculate the derivative $\partial \ln |N(q)|^2 / \partial m$. We find that for a symmetric potential (wells, of equal depth, $U_{\rm right} = 0$) we get an inverse isotope effect (negative α). This is in agreement with previous calculations[18] where the isotope effect is also derived from the coupling term. However, for our model the wells are asymmetric. As the wells become more asymmetric, α increases, approaches zero, and then changes sign with increased asymmetry of the wells, leading to the usual positive isotope effect. In Fig. 5 we show the calculated value of α for a barrier height of 20 meV and a variable right well potential $U_{\rm right}$. The asymmetry in the potential arises from the strain interaction between the tunneling units and hence is absent in earlier double-well models.[16]

As a consistency argument for our result we should mention that at about 200 K, which we choose as our barrier height, the oxygen is no longer localized in one of the wells as was found previously.[25]

One of the shortcomings of this calculation is that we do not know how the depth of the wells and, hence, how the isotope effect are related to the oxygen concentration.

B. *c*-axis tunneling

We next make some qualitative remarks about experiment on *c*-axis tunneling performed in Ref. 48. The tunneling of

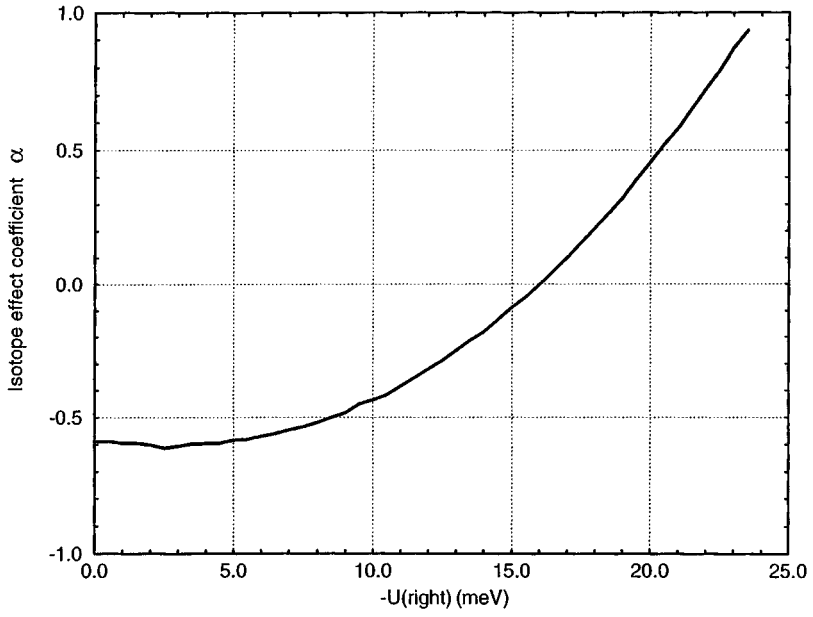

FIG. 5. Isotope effect exponent α calculated for $N_0V_1 = -1.8$ and $N_0V_2 = 1.6$. The barrier height is fixed at $U_{\rm barrier} = 20$ meV. The depth of the right well $U_{\rm right}$ is used as a parameter. See Fig. 1 for the definition of $U_{\rm barrier}$ and $U_{\rm right}$.

pairs along the c direction indicates an s-wave contribution and an almost complete absence of a d-wave contribution. Qualitatively this idea is consistent with our derivations. We get a combination of s-wave and d-wave solutions. The d waves have lobes which are 90° out of phase with each other. Thus when the pairs tunnel along the c axis the d-wave contributions will cancel (if the perpendicular lobes are of the same magnitude) and one is left with an s-wave contribution as is observed in experiments.

C. Possible other experiments

If our model is applicable to high-T_c superconductors, the the effective number of tunneling units is of the order one per unit cell. Thus an experiment that could probe the number of TU's in the high-T_c material should show this.

The coupling of the electrons to the tunneling units could be derived from a relaxation experiment similar to the one performed in glasses.[46] It would be useful to obtain this parameter which enters into Eq. (9).

X. CONCLUSION

We explore the consequences of electron scattering from interacting tunneling units (TU's) and find that it could give high-temperature superconductivity with a gap function $\Delta(\mathbf{k})$ which has a combination of s-wave and $d_{x^2-y^2}$-wave symmetries. For certain parameters we obtain nodes in the $\Delta(\mathbf{k})$. Tunneling units were found in experiments on a number of high-T_c materials. The anisotropy in $\Delta(\mathbf{k})$ arises because the TU's have a well-defined direction of orientation with respect to the crystal axis as was found in experiments. We emphasize that for our derivations of the transition temperature T_c it matters little whether the oxygen atoms or the copper atoms, or any other atoms, tunnel. As long as TU's are in a well-defined direction with respect to the crystal axis we obtain a combination of s-wave and d-wave symmetries in the gap function. However, experimental properties, for example, the isotope effect, will depend on the specific atom which tunnels. In our derivation an analogy is established between the scattering of conduction electrons by phonons in BCS theory and the scattering of electrons by TU's in our model. Because the excitation energies arising from the elastic interactions between TU's are expected to be much greater than the phonon excitation energies in BCS theory, we obtain a higher transition temperature. Thus in our model elastic excitations play a very important role and therefore we expect a strong pressure dependence of the transition temperature. We calculated the isotope effect and find that for symmetric wells we get an inverse isotope effect (negative α). However, as the wells become asymmetric because of the interactions between the tunneling units, α increases, becomes zero, and for more pronounced asymmetry becomes positive. We also comment that our model may explain why one observes only an s-wave component in c-axis tunneling of pairs.[48]

ACKNOWLEDGMENT

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APPENDIX: THE EXCITATION ENERGY OF THE TUNNELING UNITS

Derivation of the equation of motion for σ_i^x using fermion creation and annihilation operators $a_i^\dagger$ and a_i.[49] The interaction part for the system of pseudospins which represent TU's is assumed to be of the form

$$H^I=-\sum_{i<j} J_{ij}\sigma_i^z\sigma_j^z-\sum_i \xi_i\sigma_i^z \tag{A1}$$

and the tunneling part of the Hamiltonian of the form

$$H^T=-\sum_i \Delta_i\sigma_i^x, \tag{A2}$$

where σ_i^z and σ_i^x are Pauli spin matrices.

We introduce the representation of spin operators in terms of Fermi creation and annihilation operators $a_i^\dagger$ and a_i using the form suggested by Jordan and Wigner, described in the book of Mattis.[50]

We use the following definitions

$$\sigma_i^+=2a_i^\dagger Q_i, \quad \sigma_i^-=2Q_i a_i, \quad \sigma_i^z=2a_i^\dagger a_i-1, \tag{A3}$$

where

$$Q_i=Q_i^+=Q_i^{-1}=\prod_{j<i}(a_j^\dagger+a_j)(a_j^\dagger-a_j). \tag{A4}$$

The operator σ_i^x is expressed as

$$\sigma_i^x=\frac{1}{2}(\sigma_i^-+\sigma_i^+)=(Q_i a_i+a_i^\dagger Q_i). \tag{A5}$$

The Fermi operators $a_i^\dagger$ and a_i obey the commutation relations

$$\{a_i,a_j\}=0, \quad \{a_i^\dagger,a_j^\dagger\}=0, \quad \{a_i,a_j^\dagger\}=\delta_{ij}. \tag{A6}$$

Eigenvalues 0 and 1 of the Fermi number operator $a_i^\dagger a_i$ correspond to the eigenvalues of -1 and $+1$ of the operator σ_i^z

Substituting for Pauli spin operators in the expressions for the interaction and tunneling terms of the Hamiltonian [Eqs. (A1) and (A2)] we obtain

$$H^I=-\sum_{i<j} J_{ij}(2a_i^\dagger a_i-1)(2a_j^\dagger a_j-1)-\sum_i \xi_i(2a_i^\dagger a_i-1) \tag{A7}$$

and

$$H^T=-\sum_i (Q_i a_i+a_i^\dagger Q_i). \tag{A8}$$

Consider a TU at site i. As was stated in the text, at low temperature the particle in the two-level state will be found in its lowest-energy state. We denote this state as the ground state of the particle at site i, whereas when the particle tunnels to the higher-energy level it will be denoted as the excited state.

Let $|0\rangle$ denote the ground state of the system. Then for any site i

$$a_i|0\rangle = 0|0\rangle, \quad a_i^\dagger a_i|0\rangle = 0|0\rangle, \tag{A9}$$

$$\sigma_i^x|0\rangle = |i\rangle, \tag{A10}$$

where $|i\rangle$ denotes the state with ith fermion excited.

We use the relation $Q_i|0\rangle = |0\rangle$ and we let E_0 and E_1 be the ground-state and excited-state energies, respectively. We then obtain

$$[\sigma_i^x, H^I]|0\rangle = (E_0 - E_i)|i\rangle = -\left(2\sum_j J_{ij} - \xi_i\right) a_i^\dagger|0\rangle. \tag{A11}$$

We finally have

$$E_i - E_0 = \left(2\sum_j J_{ij} - \xi_i\right). \tag{A12}$$

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