On superconductivity in systems with non-linear electron–phonon coupling and structural instabilities

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Abstract. A model for the linear and quadratic coupling of an anharmonic optical phonon branch producing a structural phase transition at $T = T_0$ with a band of free electrons has been treated within an extended Eliashberg theory. The quadratic coupling causes a critical behaviour of $T_c(T_0)$ as $T_c \to T_0$ due to a renormalization of the linear coupling constant by a non-vanishing lattice order parameter for $T < T_0$. The parameter of the isotope effect α shows an unusual behaviour. Zero or even negative values of α are obtained if the superconducting transition is near the structural one.

1. Introduction

Up to now there has been no complete theoretical study on the influence of anharmonicity in the lattice potentials on the superconducting transition temperature T_c and the isotope effect for electron–phonon-interaction-driven superconductivity. Problems appear in defining and evaluating the elementary excitations of lattices which are characterized by large thermal amplitudes of ionic vibrations and hence by strong anharmonicity. The effect of anharmonicity can be discussed on the basis of the following on-site potential for a local normal coordinate Q of some lattice mode: $V(Q) = aQ^2 + bQ^4$. Two cases have to be considered.

If $a > 0$ then the anharmonicity becomes significant only for high enough temperatures that $a/b \le \langle Q^2 \rangle$ holds. Then the temperature dependence of the phonon frequencies and of the phonon lifetime (see [1]) can cause a substantial temperature dependence of the Eliashberg function. For low enough temperatures a perturbation approach works. As was shown in [2] the excitation spectra and consequently the superconducting properties will be changed only slightly compared with the case of harmonic phonons. However, the effect of phonon scattering (line broadening) will reduce T_c, as was demonstrated in [3]. If the parameter a becomes negative we get a double-well potential. In such a case the lattice system can exhibit a structural phase transition (SPT) [4]. The influence of this on the superconductivity was analysed in [5]. There it was assumed that the sum of stabilizing long range forces exceeds the local harmonic instability a. At the SPT point T_0 the excitation energies of the lattice under consideration go to zero. This soft mode behaviour leads to a strong correlation of T_c and T_0. The maximum value of T_c is reached if the lattice is most unstable, i.e. if the lattice parameters lead to a SPT temperature which is equal to T_c.

The second consequence of a soft mode is a strong variation of the isotope effect of T_c. For a certain parameter range the isotope effect becomes very small or even negative. Another interesting case was discussed several years ago in [6]. Here the motion in the double-well potential is considered for the strong low-temperature quantum case of coupled two-level systems. A remarkable increase of T_c due to the large thermal fluctuations of the lattice motion was found. The isotope effect for such a model was investigated in [7] and positive or negative values depending on λ were found.

As we are dealing with lattices showing large thermal amplitudes of ions due to anharmonic motions it is certainly necessary to take into account electron–lattice coupling not only linearly in the displacements but at least also quadratically. The influence of non-linear coupling was already discussed in [3] and [8]. There are two effects. For small fluctuations [3] it was shown that the coupling constant λ is reduced due to the Debye–Waller factor. For strong fluctuations, however, an increase of T_c was predicted by [8]. Considering certain modes such as the tilting mode in the La_2CuO_4 system, the electron–lattice coupling quadratic in the displacements plays the central role, as the linear coupling term is absent for reasons of symmetry (see [9]). A different approach to the effect of anharmonicity on superconductivity was discussed in the series of papers [10], [11], [12]. A double-shell model, e.g. for the oxygen ions, with coupled double-well potentials combines the ferroelectric instability due to an anharmonic electron–phonon interaction with an electronic instability. As regards dependence on the model parameters, either a transition into a ferroelectric or into a superconducting state can be described within a self-consistent phonon approximation. The isotope effect of T_c was found to be much smaller than for the BCS theory.

The aim of this paper is to reconsider the influence of modes which are of special symmetry and connected with some kind of lattice instability. This will be done in a schematic model (section 2). For this model the Eliashberg theory is developed (section 3), the lattice properties including the two-phonon Green function are calculated (section 4) and the results are compared with previous ones without quadratic coupling (section 5).

2. The model and Eliashberg theory for linear and quadratic coupling

As we are interested in discussing the general features of the influence of anharmonicity on the superconductivity we will treat a schematic model and do not claim to describe any specific experimental situation in high-T_c materials. However, for numerical estimations we use parameters typical for high-T_c superconductors. The model has the following form:

$$H = H_{el} + H_{int} + H_{lattice} \qquad (2.1)$$

where H_{el} describes a simple band in Wannier representation:

$$H_{el} = \sum_{ij\sigma} t_{ij} c_{i\sigma}^{+} c_{j\sigma}. \qquad (2.2)$$

$H_{lattice}$ stands for the lattice subsystem containing only one vibration branch causing a SPT:

$$H_{lattice} = \sum_{i} \left(\frac{p_i^2}{2m} - \frac{A}{2} x_i^2 + \frac{B}{4} x_i^4 \right) + \frac{1}{4} \sum_{ij} C_{ij} (x_i - x_j)^2. \qquad (2.3)$$

For the interaction of the lattice vibrations with the electron band we take into account linear and quadratic coupling in a simple form:

$$H_{\text{int}} = \sum_i p\,x_i\,c_i^+ c_i + \sum_i q\,x_i^2\,c_i^+ c_i. \tag{2.4}$$

For a convenient treatment the Hamiltonian (2.1) was rewritten in the Nambu matrix formalism (see e.g. [13])

$$H = \sum_{ij} t_{ij}(c_i\,{}^+\tau_3 c_j) + \sum_i p x_i(c_i\,{}^+\tau_3 c_i) + \sum_i q\,x_i^2\,(c_i\,{}^+\tau_3 c_i) + H_{\text{lattice}} \tag{2.5}$$

where $c_i\,{}^+$, c_i are electron vector operators:

$$c_i = \begin{bmatrix} c_{i\uparrow} \\ c_{i\downarrow}^+ \end{bmatrix} \qquad c_i\,{}^+ = [c_{i\uparrow}^+ c_{i\downarrow}]. \tag{2.6}$$

The $\tau_i (i = 0, 1, 2, 3)$ are the Pauli matrices. The one-electron matrix Green function

$$\hat{G}_{ij}(\omega) = \langle\!\langle c_i | c_j\,{}^+\rangle\!\rangle = \begin{bmatrix} \langle\!\langle c_{i\uparrow} | c_{j\uparrow}^+ \rangle\!\rangle & \langle\!\langle c_{i\uparrow} | c_{j\downarrow} \rangle\!\rangle \\ \langle\!\langle c_{i\downarrow}^+ | c_{j\uparrow}^+ \rangle\!\rangle & \langle\!\langle c_{i\downarrow}^+ | c_{j\downarrow} \rangle\!\rangle \end{bmatrix} \tag{2.7}$$

(with $\langle\!\langle A(t) | B(t') \rangle\!\rangle = -i\Theta(t - t')\langle [A(t), B(t')]_\pm \rangle$, $\langle \ldots \rangle$ indicating thermodynamical averaging, and $A(t)$ Heisenberg representation of the operator A) is calculated with the equation of motion method. This leads to an equation $\hat{G} = \hat{G}_0 + \hat{G}_0 \hat{P} \hat{G}_0$ where the zeroth-order GF is given by

$$\hat{G}_k^0(\omega) = (\omega\tau_0 - \epsilon_k\tau_3)^{-1} \qquad \epsilon_k = t_k + \tilde{p}_i\langle x_i\rangle + q\langle u_i^2\rangle$$
$$u_i = x_i - \langle x_i\rangle \qquad \tilde{p}_i = p + 2q\langle x_i\rangle \qquad \eta = \langle x_i\rangle. \tag{2.8}$$

The renormalization of the linear electron–phonon coupling constant p in (2.8) due to non-vanishing static displacements becomes significant if the lattice undergoes a SPT, since $\langle x_i\rangle$ is temperature dependent (see section 3 and figure 2). In the case of vanishing electron–phonon coupling (isolated lattice) two solutions with $\langle x_i\rangle = \pm|\langle x_i\rangle|$ can appear at the SPT. Switching on the electron–phonon coupling, the eigenvalues ϵ_n of the Hamiltonian (2.1) exhibit the symmetry $\epsilon_n(x_i) = \epsilon_n(-x_i)$ in a perturbation approach if the high-symmetry phase is assumed. However, this symmetry is broken if the SPT is reached and $q \neq 0$ is assumed, since different $\tilde{p}$ (see (2.8)) lead to different reductions of the ϵ_n within the conventional perturbation approach, i.e. larger $\tilde{p}$ cause lower ϵ_n. Thus larger $\tilde{p}$-values are favoured.

For the polarization operator $\hat{P}$ the following expression is valid:

$$\hat{P}_{ij}(\omega) = \tau_3 \left(\tilde{p}^2 \langle\!\langle c_i u_i | c_j^+ u_j \rangle\!\rangle + q^2 \langle\!\langle c_i \delta u_i^2 | c_j^+ \delta u_j^2 \rangle\!\rangle \right) \tau_3 \qquad \delta u_i^2 = u_i^2 - \langle u_i^2\rangle. \tag{2.9}$$

The Dyson equation $\hat{G} = \hat{G}_0 + \hat{G}_0 \hat{M} \hat{G}$ provides the relation between $\hat{P}$ and the mass operator $\hat{M}$ of the electron GF:

$$\hat{M} = \hat{P}(1 + \hat{P}\hat{G}_0)^{-1} = \hat{P}^{\text{irr}}. \tag{2.10}$$

The superscript 'irr' denotes the irreducible part (with respect to $\dot{G}_0$) of the GF under consideration. Decoupling electron and phonon variables in $\dot{M}$ an expression can be found which coincides with the corresponding formula in the usual Eliashberg theory with the only difference that beside the one-phonon GF $D_{ij}^{(1)}(\omega) = \langle\langle u_i|u_j\rangle\rangle$ we have the two-phonon GF $D_{ij}^{(2)}(\omega) = \langle\langle u_i^2|u_j^2\rangle\rangle$. Consequently the apparatus of the Eliashberg theory has been applied in the usual manner. For the Eliashberg function $\alpha^2F(\omega)$ we get:

$$\alpha^2F(\omega) = \frac{N(E_F)}{\pi\hbar N}\left(\bar{p}^2\sum_{k}\mathrm{Im}D_k^{(1)}(\omega) + q^2\sum_{k}\mathrm{Im}D_k^{(2)}(\omega)\right) \tag{2.11}$$

where $N(E_F)$ is the electron density of states at the Fermi energy E_F. To calculate T_c we use a standard interpolation formula derived by Krezin [14]:

$$kT_c = \frac{0.25\hbar\Omega}{\sqrt{e^{\frac{2}{\lambda}} - 1}} \tag{2.12}$$

where the electron–phonon coupling constant λ and the averaged phonon frequency Ω are given by:

$$\lambda = \int_{-\infty}^{\infty} d\omega\, \alpha^2F(\omega)/\omega \tag{2.13}$$

$$\Omega^2 = \frac{1}{\lambda}\int_{-\infty}^{\infty} d\omega\, \alpha^2F(\omega)\omega. \tag{2.14}$$

3. Treatment of the anharmonic lattice

The lattice Hamiltonian H_{lattice} describes a system with a SPT of both displacive and order–disorder type depending on the ratio A/C_0 where $C_0 = \sum_{i\neq j} C_{ij}$ is the integrated interaction strength (see e.g. [15]). Here we are interested in a parameter range $(A/C_0 < 1)$ where a so-called soft mode behaviour occurs, i.e. where an optical phonon frequency $\omega_q(T)$ tends to zero for a certain wave vector q_c as the SPT temperature T_0 is reached $(\omega_{q_c}(T = T_0) = 0)$ [4]. For $T < T_0$ an order parameter η is involved. η is connected with static displacements of the ions $\langle x_i\rangle$:

$$\langle x_i\rangle = \eta\, e^{iq_c\cdot R_i}. \tag{3.1}$$

R_i is the position vector of the ith elementary cell. For sake of simplicity we assume long range interaction forces $C_{ij} = C_0/N$ (N = number of elementary cells) and hence $q_c = 0$. This assumption does not change the general features of the phonon system which are essential for our consideration namely a soft mode behaviour and a SPT. To treat the anharmonicity x^4 in H_{lattice} we use the standard Green function technique. The thermodynamical averages are carried out with H_{lattice} only, i.e. the features of the phonon system are calculated neglecting the electron–phonon interaction as is usually done in the framework of the Eliashberg theory [13]. To evaluate the phonon Green function $D^{(1)}(\omega)$ we use the self-consistent phonon approximation for the decoupling

of correlation functions of higher order which works well if $A/C_0 < 1$ (see [15]), i.e. in the parameter range of interest. This leads to the following coupled set of equations:

$$D_q^{(1)}(\omega) = \frac{\hbar}{m(\omega^2 - \omega_q^2)} \tag{3.2}$$

$$m\omega_q^2 = \Delta^2 + C_0 - C_q \tag{3.3}$$

$$\Delta^2 = -A + 3B\langle u_i^2 \rangle + 3B\eta^2 \tag{3.4}$$

$$\eta\left(-1 + \eta^2 + 3\langle u_i^2 \rangle\right) = 0 \tag{3.5}$$

$$\langle u_i^2 \rangle = \frac{\hbar}{2mN} \sum_q \frac{1}{\omega_q} \coth\left(\frac{\hbar\omega_q}{2kT}\right). \tag{3.6}$$

The temperature of the SPT, T_0, is given by

$$kT_0 = \frac{\hbar\sqrt{C_0}}{2\sqrt{m}\,\coth^{-1}\left(\frac{2\sqrt{C_0 m A}}{3B\hbar}\right)}. \tag{3.7}$$

Within the same order of approximation (self-consistent phonons, i.e. non-interacting quasiparticles with temperature dependent energy) the two-phonon GF can be expressed in terms of the one-phonon GF:

$$
\begin{aligned}
D_{ij}^{(2)}(\omega) = \langle\langle u_i^2 | u_j^2 \rangle\rangle &= \frac{1}{2\pi} \int d\bar{\omega} \frac{e^{\beta\bar{\omega}} - 1}{\omega - \bar{\omega}} \int dt\, e^{-i\bar{\omega}t} \langle u_j^2(t) u_i^2 \rangle \\
&\approx \frac{2}{\pi} \int d\bar{\omega} \frac{e^{\beta\bar{\omega}} - 1}{\omega - \bar{\omega}} \langle u_j(t) u_i \rangle \langle u_j(t) u_i \rangle \\
&= \frac{2}{\pi^2} \int d\bar{\omega}\, d\bar{\bar{\omega}} \frac{e^{\beta\bar{\omega}} - 1}{\omega - \bar{\omega}} \frac{\mathrm{Im}D_{ij}^{(1)}(\bar{\bar{\omega}})}{e^{\beta\bar{\bar{\omega}}} - 1} \frac{\mathrm{Im}D_{ij}^{(1)}(\bar{\omega} - \bar{\bar{\omega}})}{e^{\beta(\bar{\omega} - \bar{\bar{\omega}})} - 1}.
\end{aligned} \tag{3.8}
$$

With the dispersion relation chosen above and inserting (3.2) and (3.6) we obtain

$$D_{ij}^{(2)}(\omega) = \frac{4\hbar\langle u_i^2 \rangle}{m(\omega^2 - 4\omega_0^2)}\delta_{ij} \tag{3.9}$$

with

$$\omega_0^2 = \Delta^2 + C_0. \tag{3.10}$$

4. Numerical results and discussion

With the information gained in the preceding section the Eliashberg function (cf. equation (2.11)) is given

$$\alpha^2 F(\omega) = \frac{N(E_{\mathrm{F}})}{m\omega_0}[\bar{p}^2\,(\delta(\omega - \omega_0) - \delta(\omega + \omega_0)) + q^2\,\langle u_i^2 \rangle\,(\delta(\omega - 2\omega_0) - \delta(\omega + 2\omega_0))]. \tag{4.1}$$

Inserting this result into (2.13), (2.14) gives

$$\lambda = \frac{N(E_F)}{m\omega_0^2}(\bar{p}^2 + q^2\langle u_i^2\rangle) \tag{4.2}$$

$$\Omega^2 = \frac{N(E_F)}{\lambda m}(\bar{p}^2 + 4q^2\langle u_i^2\rangle). \tag{4.3}$$

Because of the T-dependence of ω_0 and $\langle u^2\rangle$ (see equations (3.6), (3.10)) λ and Ω become T-dependent ((4.2), (4.3)). To determine T_c one has to solve (3.3), (3.5), (3.6), (2.12) self-consistently.

In order to get reasonable values for the model parameters A, B, C_0 and m we consider the tetragonal–orthorhombic SPT in $La_{2-x}Sr_xCuO_4$. This SPT is caused by the softening of the tilting mode (rigid rotations of CuO_{6-} octahedra) at the X point of the Brillouin zone [16]. A microscopical model for this SPT was given in [17], [18]. Plakida [19] transformed this model into a ϕ^4-model (2.3). The Sr-doping x was expected to influence mainly the harmonic instability A due to changes of the ionic charges at La(Sr) positions and the appearance of free charge carriers [17], [18]. Since for $x = 0$ $T_0(La_2CuO_4) = 530$ K is measured [20] the high-temperature limit of equation (3.7) can be considered: $T_0 \sim A$. From the experimental result $T_0 \sim (1 - 4x)$ [20] it follows that $A = A_0(1 - 4x)$. An estimation of the model parameters was given in [5], [17] using neutron scattering results:

$$\frac{\hbar^2 A_0}{m} \approx 60 \text{ meV}^2 \qquad \frac{A_0}{B} \approx 0.09 \text{ Å}^2 \tag{4.4}$$

$$\frac{C_0}{A_0} \approx 2.44 \qquad m \approx 2.6 m_{oxygen}. \tag{4.5}$$

The electron density of states was assumed as $N(E_F) = 1 \text{ eV}^{-1}$ [21]. Note that generally x can be considered as a control parameter for shifting the SPT temperature T_0 and thereby the correlations between T_0 and T_c can be studied.
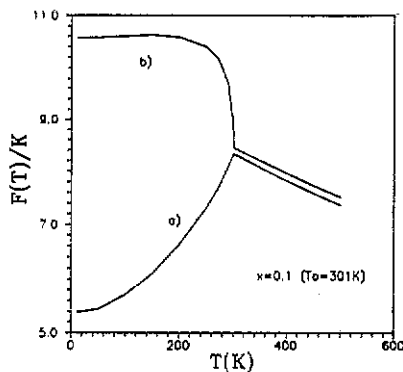

Figure 1. Right-hand side of (2.12) as function of temperature for (a) purely linear coupling ($p = 1 \text{ eV Å}^{-1}$); (b) linear + quadratic coupling ($p = 1 \text{ eV Å}^{-1}, q = 0.5 \text{ eV Å}^{-2}$).
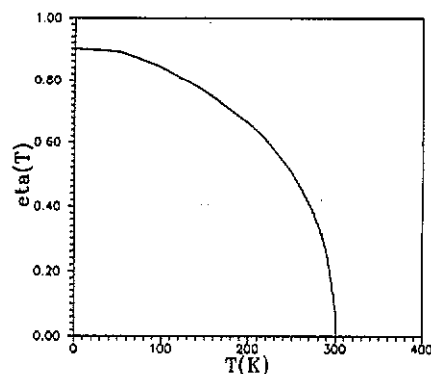
Figure 2. Lattice order parameter $\eta(T)$ for $x = 0.1$ (corresponds to $T_0 = 301$ K).

The features of the above discussed model in the case $q = 0$ (only linear electron–phonon coupling) were given in [5], where a p-value of 1 eV $Å^{-1}$ was used. Here we want to study additional effects of quadratic coupling $(q \neq 0)$. In a first step of solving our system of equations we determined $F(T)$ for $x = 0.1$ and $q = 0.5$ eV $Å^{-2}$ where $F(T)$ is the right-hand side of (2.12) as a function of temperature (figure 1). For comparison we show our recent results for $q = 0$ (cf. [5]). The main influence of quadratic electron–phonon coupling in this parameter range is mediated by the renormalization of the linear term p due to the appearance of an order parameter $\eta(T)$ (figure 2). While for $q = 0$ we got a maximum in $F(T)$ at T_0 due to the softening of the phonon frequency ω_0 at the SPT, now the rapid increasing of η below T_0 ($\eta \sim (T_0 - T)^{1/2}$) causes a drastic enlargement of $F(T)$ below T_0. However, in the high-symmetry phase ($\eta = 0$) the bare additional influence of quadratic electron–phonon coupling (see (4.2), (4.3)) is negligibly small. This fact holds for q-values up to 10 eV $Å^{-2}$.
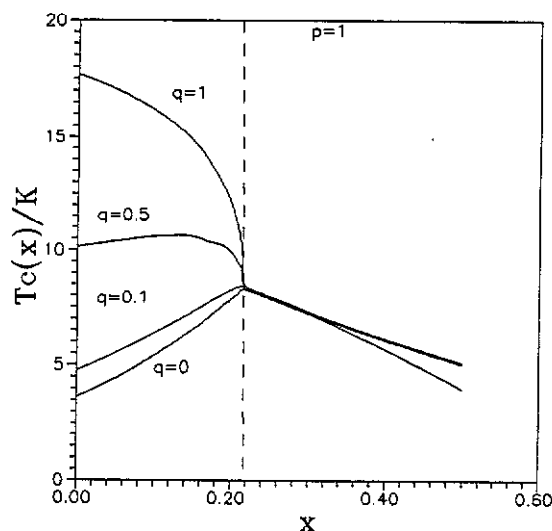
Figure 3. Superconducting transition temperature T_c as function of the control parameter x, which shifts the SPT temperature, for different quadratic coupling strengths q (in eV $Å^{-2}$).

Now let us calculate $T_c(x)$ by solving the self-consistent equation $T = F(T)$. The results are shown in figure 3. With increasing q the maximum in $T_c(x)$ shifts from the x-value where $T_c = T_0(q = 0)$ to $x = 0$ ($q \geq 1$ eV $Å^{-2}$). The critical behaviour of $T_c(x)$ for $x \to x_c$ is due to the fact that at x_c the SPT appears and the critical behaviour of $\eta(x)$ leads to a similar one of $\tilde{p}(x)$ (see (2.8)). Thus for $q = 0.5$ eV $Å^{-2}$ a dependence can be reproduced where $T_c(x)$ is nearly constant in the low-symmetry phase ($\eta \neq 0$) and rapidly decreases as the high-symmetry phase ($\eta = 0$) is reached. The parameter of the isotope effect $\alpha = d(\ln T_c)/ d(\ln m)$ exhibits suprising features within our model. To calculate the dependence of α on x for the above model parameters we determined T_c for O^{16} and for the O^{18} oxygen isotope mass and then α according to the relation

$$\alpha = \frac{\Delta T_c}{T_c(O^{16})} \frac{m(O^{16})}{\Delta m} \tag{4.6}$$

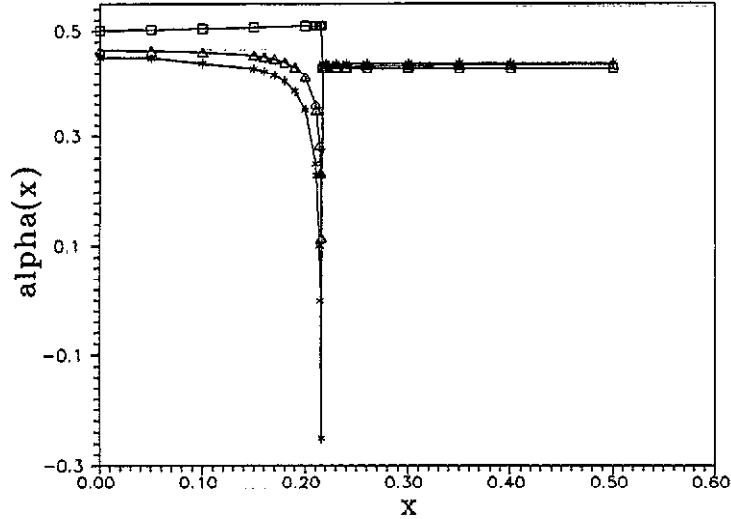$$\Delta T_c = T_c(O^{16}) - T_c(O^{18}) \qquad \Delta m = m(O^{18}) - m(O^{16}). \tag{4.7}$$

Figure 4. The parameter of the isotope effect α as a function of the control parameter x for $q = 0(\square), q = 0.5$ eV $\text{Å}^{-2}(\triangle)$ and $q = 1$ eV $\text{Å}^{-2}(*)$.
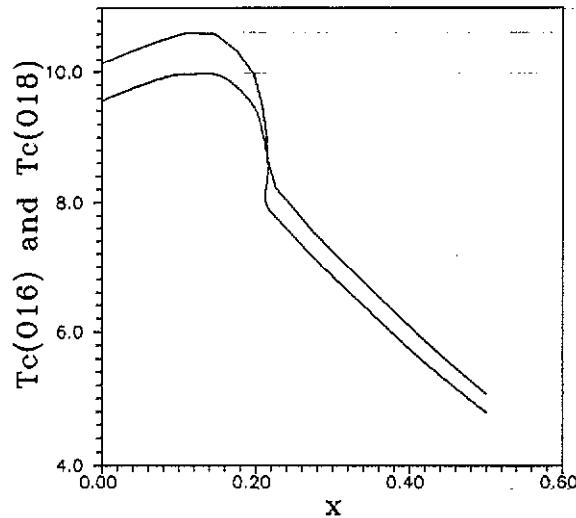

Figure 5. Shift of T_c due to isotope mass change, $p = 1$ eV $\text{Å}^{-1}, q = 0.5$ eV Å^{-2}.

The $\alpha(x)$-dependence is shown in figure 4. For $q = 0\alpha(x)$ shows a steplike decrease ($T_c = T_0$) while $\alpha(x)$ is nearly independent of x for $x < x_c$ and $x > x_c$. The value of α is nearly the same as for BCS superconductors: $0.43 \leq \alpha \leq 0.5$. With increasing q strong deviations from the $q = 0$ case appear near x_c if $x < x_c$. α decreases drastically for $x \rightarrow x_c$ and even reaches negative values for strong enough quadratic coupling. This behaviour is a consequence of the dependence of T_c on x (see figure 5). For slightly enlarged isotope mass the $T_c(x)$-curve shifts downward and to higher x-values.

Let us discuss the results. The main effect of quadratic electron–phonon interaction on T_c comes through the renormalization of the linear coupling term p due to non-vanishing static ion displacements. Such temperature dependent displacements occur

if the lattice undergoes a SPT. That means that the expansion of the electron hopping matrix elements in powers of the lattice displacements around the equilibrium positions of the ions lead to a temperature dependent linear electron–phonon matrix element p. Thus one has to be careful to use $\alpha^2 F(\omega)$-data from experiments done at higher temperatures (e.g. room temperature) than T_c or even T_0 since the possible existence of a SPT can change the electron–phonon coupling crucially due to an increasing order parameter with lowering temperature. Especially in the case of high-temperature superconductors ($T_c \geq 100$ K) it seems to be possible that SPT at lower temperatures, i.e. in the superconducting state, can appear and cause a temperature dependent renormalization of the electron–phonon coupling. Then the temperature dependence of the superconducting gap and the critical magnetic field is changed and the ratio $2\Delta/kT_c$ becomes larger than the BCS-value.

5. Summary

In the present work we treated a simple model which describes free electrons coupled to anharmonic lattice vibrations linear and quadratic in the displacements. The lattice undergoes a structural phase transition at T_0 with a soft mode behaviour. While with purely linear coupling the superconducting transition temperature $T_c(T_0)$ has its maximum if $T_c = T_0$, non-vanishing quadratic coupling causes a critical behaviour of $T_c(T_0)$ as $T_c \to T_0$. This is due to a strong renormalization of the linear coupling constant by the existence of an order parameter for $T < T_0$. At the same time the isotope effect rapidly decreases as $T_c \to T_0$ and even zero or negative values are obtained.

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