Modification of the isotope effect due to energy-dependent electronic density of states

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We study how energy dependence in the electronic density of states $N(\varepsilon)$ affects the isotope-effect coefficient β within a realistic Eliashberg formalism. For structure in $N(\varepsilon)$, sharp on the scale of the Debye energy, the value of β can be significantly affected and can be smaller or larger than the canonical value 0.5. In some cases, when coupled with large values of the Coulomb pseudopotential μ^* , β can be quite small with otherwise fairly unremarkable values of the other parameters.

I. INTRODUCTION

The value of the isotope coefficient $\beta = -d \ln(T_c)/d$ $d \ln(M)$, where T_c is the critical temperature and M is the ion mass, has played a significant role in our understanding of the mechanism responsible for Cooper pairing and consequently superconductivity. A value $\beta = 0.5$ is predicted for an electron-phonon mechanism within BCS theory, while smaller values are possible in Eliashberg theory when the competition between the Coulomb pseudopotential μ^* and electron-phonon mass renormalization λ is important. Roughly comparable values of these two parameters can occur when T_c is small and a zero or even negative isotope-effect coefficient can result. On the other hand for $T_c \gtrsim 10$ K, β is expected, in most cases, to be only slightly less than 0.5 because λ is very large compared with the canonical value of μ^* , which is approximately 0.1.

In conventional Eliashberg theory, the electronic density of states $N(\varepsilon)$ is assumed not to vary in the energy range around the Fermi energy ε_F of importance for superconductivity, which is the phonon energy ω_E . In this case, it can be fixed to its value N(0) at $\varepsilon_F=0$. On the other hand, if $N(\varepsilon)$ does vary on the scale of ω_E it is some average of $N(\varepsilon)$ over the range $(-\omega_E, \omega_E)$ from ε_F that determines T_c .¹⁻⁷ But, when ω_E is changed, as in the isotope effect, this average will also change and β will correspondingly be affected.⁶ In general this change in the average density of states can be an increase or a decrease and so T_c can go up or down.

In this paper we use a generalized Eliashberg formalism⁸⁻¹² with modifications to include the full energy dependence of $N(\varepsilon)$. Before going into the full complications needed to do a realistic calculation we start, in Sec. II, with the simple BCS result because it is analytic and turns out to be qualitatively correct. In Sec. III we present the necessary generalized Eliashberg equations and give preliminary results for parameters that, at least in one case, should be realistic for Nb₃Sn. Variations in $N(\varepsilon)$ that are more pronounced than those presently indicated by band-structure calculations and experiment are also considered.¹³⁻¹⁹ Additional calculations with fairly unconstrainted parameters for a T_c value around 100 K are also presented with a view to exploring the range of results that are possible when we allow fairly free ranging parameters.

In Sec. IV we present results of a generalization, to include energy dependence in $N(\varepsilon)$, of the original work of Rainer and Culetto²⁰ on the differential isotope effect coefficient $\beta(\omega)$, which gives the contribution to the total isotope effect of a particular phonon frequency ω . The coefficient $\beta(\omega)$ is found to reflect the main features of the functional derivative $\delta T_c / \delta \alpha^2 F(\omega)$ and helps us understand how energy dependence in $N(\varepsilon)$ can modify $\beta(\omega)$ and by implication the total isotope coefficient $\beta = \int_0^\infty d\omega \beta(\omega)$. To illustrate the point further, partial isotope effects are considered in a simple two δ function model for the electron-phonon spectral density.

II. A SIMPLE BCS APPROACH

To make our main points it will be sufficient to use for the single-spin electronic density of states around the Fermi energy a Lorentzian form of width a, superimposed on a constant background N_b with the Fermi energy falling a distance b from the center of the Lorentzian peak. Thus, for $N(\varepsilon)$ we have

$$N(\varepsilon) = N_b \left[1 + \frac{g}{\pi} \frac{a}{a^2 + (\varepsilon + b)^2} \right], \qquad (1)$$

where $\varepsilon = 0$ gives the Fermi level, and $q/(\pi a)$ is the height of the Lorentzian above the background that occurs at $\varepsilon = -b$. At the Fermi level this height is reduced by a factor of $a^2/(a^2+b^2)$ and is denoted *D*. In a BCS model, the equation for T_c can be solved analytically²¹ and the isotope coefficient β follows directly:

$$\beta = \left| \frac{d \ln T_c}{d \ln M} \right|$$

$$= \frac{1}{2} - \left(\frac{D}{1+D} \right) \frac{\omega_D}{4} \left[\frac{\omega_D - 2b}{a^2 + (\omega_D - b)^2} + \frac{\omega_D + 2b}{a^2 + (\omega_D + b)^2} \right]. \quad (2)$$

In Fig. 1 we show results for β as a function of b/a for five values of a/ω_D , namely, 0.2 (solid line), 0.4 (dotted

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FIG. 1. The isotope coefficient β as a function of b/a given by Eq. (2) in BCS theory. The values of a/ω_D are 0.2 (solid), 0.4 (dotted), 0.6 (medium dashed), 0.8 (long dashed), and 1.0 (dot dashed). In all curves D is fixed at $5/\pi$.

line), 0.6 (medium-dashed line), 0.8 (long-dashed line), and 1.0 (dot-dashed line). We note that for small b and $a = 0.2\omega_D$, β can be as small as 0.2, while for the same value of a it can be larger than 0.8 when $b/a \approx 5.0$. As b becomes large compared with the width of the Lorentzian, β approaches the classical BCS value of 0.5 because we are then so far away from the Lorentzian peak that it plays little role, and we get the same result as for a constant density of states.

Differentiation of the square brackets in Eq. (2) gives a minimum at b = 0, in which case β takes on a particularly simple form, namely,

$$\beta = \frac{1}{2} \left[1 - \frac{D}{1+D} \frac{1}{1+\overline{a}^2} \right], \qquad (3)$$

where $\bar{a} = a/\omega_D$. For a positive value of D this equation gives minimum β when $\bar{a} = 0$, in which case $\beta = 1/[2(1+D)]$, which goes to zero like 1/D as $D \to \infty$. Therefore, in BCS theory, an infinitely sharp peak of infinite height is required to get $\beta = 0$ and negative values are not possible. For this to occur we need to use Eliashberg theory.

III. FORMALISM AND RESULTS FOR Nb₃Sn

For the model of the electronic density of states given by Eq. (1) the linearized Eliashberg gap equations that give the critical temperature $\operatorname{are}^{2-5,9-11}$

$$\Delta(i\omega_n)Z(i\omega_n) = \pi T \sum_m [\lambda(n-m) - \mu^* \Theta(\omega_c - |\omega_m|)] \frac{\Delta(i\omega_m)}{|\omega_m|} \hat{N}_m$$

(4)

and

$$\omega_n Z(i\omega_n) = \omega_n + \pi T \sum_m \lambda(n-m) \operatorname{sgn}(\omega_m) \hat{N}_m$$
 (5)

with

$$\hat{N}_{n} = N_{b} \left[1 + \frac{g}{\pi} \frac{Z(i\omega_{n})|\omega_{n}| + a}{\pi [Z(i\omega_{n})|\omega_{n}| + a]^{2} + b^{2}} \right], \quad (6)$$

where $\Delta(i\omega_n)$ and $Z(i\omega_n)$ are the gap and renormalization functions evaluated at the Matsubara frequencies $i\omega_n = i(2n+1)\pi T$ with $n = 0, \pm 1, \pm 2, \ldots$, and T is the temperature. The function $\lambda(n-m)$ in Eqs. (4) and (5) is determined by the electron-phonon spectral density $\alpha^2 F(\omega)$ with

$$\lambda(n-m) = 2 \int_0^\infty d\omega \frac{\omega \alpha^2 F(\omega)}{\omega^2 + (\omega_n - \omega_m)^2} .$$
 (7)

Finally, μ^* is the Coulomb pseudopotential for a cutoff ω_c .

A first set of solutions for β were obtained for parameters similar to those that might apply to Nb₃Sn. The spectral density is taken from tunneling inversion on Nb_3Sn and the T_c set at 18.05 K. The Lorentzian form is fixed at g = 100 meV with the parameters a and b variable. For each choice of a and b, the Coulomb psuedopotential $\mu^*(\omega_c)$ was readjusted to keep the T_c value fixed and the background density of states was taken to be absorbed into the measured $\alpha^2 F(\omega)$ so that N_b becomes a dimensionless number equal to 1.0. The resulting total isotope effect coefficient β is displayed in the top frame of Fig. 2. The solid, dotted, medium-dashed and longdashed curves apply, respectively, to a = 0, $0.1\omega_D$, $0.5\omega_D$, and $1.0\omega_D$, where ω_D is the Debye energy, which is ~29 meV for Nb₃Sn. We see that for small ($\ll \omega_D$) b, β can be strongly depressed over its value when b is larger ($\gg \omega_D$). The depression is largest for $a \rightarrow 0$ but is still significant when $a = 0.1\omega_D$. As b increases, the isotope effect coefficient also increases steadily and can be larger than 0.5 in the intermediate b region $b/\omega_p \sim (1-2)$ especially for small a. A first conclusion that can be drawn is that β is strongly affected when the scale for significant energy variations in $N(\varepsilon)$ is of the order ω_D . Also, β can increase as well as decrease. It should be noted, however, that part of the change in β shown in Fig. 2 is due to the different values of $\mu^*(\omega_c)$ used in the different runs. This was necessary in order to retain a constant T_c value and is shown in the bottom frame of Fig. 2. This change in μ^* is not, however, the overwhelming effect of the introduction of $N(\varepsilon)$ into the theory and could not, for example, by itself increase β above 0.5.

We note that our plots of the Coulomb pseudopotential $\mu^*(\omega_E)$ are for a cutoff of ω_E . In actual numerical calculations we instead use a cutoff of $6\omega_E = \omega_c$ and the corresponding values of the Coulomb pseudopotential $\mu^*(\omega_c)$ is related to $\mu^*(\omega_E)$ by the Morel-Anderson²² relation

$$\mu^*(\omega_E) = \frac{\mu^*(\omega_c)}{1 + \mu^*(\omega_c) \ln(\omega_c / \omega_E)} .$$
(8)



FIG. 2. The total isotope effect for a superconductor with electronic density of state $N(\varepsilon)$ given by Eq. (1). The Lorentzian form superimposed on a constant background of height $N_b = 1$, is given by g = 100 meV, with a = 0 (solid curve), $a = 0.1\omega_d$ (dotted curve), $a = 0.5\omega_D$ (medium-dashed curve), and $a = \omega_D$ (long-dashed curve); ω_D is the Debye energy. The top frame applies to β vs b/ω_D , while the bottom frame is for $\mu^*(\omega_E)$ vs b/ω_D with β and $\mu^*(\omega_E)$ the isotope effect coefficient and Coulomb pseudopotential, respectively. The electron-phonon spectral function used was that for Nb₃Sn, and T_c is 18.05 K.

Band-structure calculations in Nb₃Sn (Refs. 13-18) have revealed the possibility that $N(\varepsilon)$ might be varying on a scale of a few tens of meV's and a reasonable, rough model for this variation, using a Lorentzian form, is $g = 85 \text{ meV}, a \approx 15 \text{ meV}, b \approx 14 \text{ meV}, and the dimension$ less background density $N_h \approx 0.7$, which has the effect of reducing the constant density-of-states factor absorbed into $\alpha^2 F(\omega)$, while, at the same time, including an additional energy-dependent part as in Eq. (1). This case is sufficiently close to the model used in Fig. 2 that we can consider the run with $a = b \approx 0.5 \omega_D$ fairly representative of real Nb₃Sn. It is clear that a realistic $N(\varepsilon)$ can indeed significantly modify β as compared to a constant N(0)calculation. From the experimental point of view, however, Nb₃Sn is a poor case, since niobium does not have any suitable stable isotopes and the tin isotope effect is expected and found to be small.²³

As a second illustrative and suggestive, but not necessarily as realistic an example, we consider the case $\mu^*=0$ and T_c values representative of the oxide superconductors. For the spectral density we take a δ function of weight A centered around the Einstein frequency ω_E , which we take to be 90 meV. This corresponds to the top of the generalized phonon frequency distribution measured by incoherent inelastic neutron scattering in Y-Ba-Cu-O.²⁴ For the density of states we use g = 700 meV with a background density N_b of 0.5 and vary both of the additional parameters a and b. In this model the Lorentzian height can get quite large. As an example for $a=0.25\omega_E=22.5$ meV, $g/\pi a$ is about 8 for $T_c=92$ K. The existence of such a large and sharp peak is not supported in present band-structure calculations for Y-Ba-Cu-O (Refs. 25-30) even though one might expect sharp structure to result due to van Hove singularities in two dimensions such as in the CuO planes. This shortcoming, however, is not a primary concern here, since we have chosen parameters by design to illustrate the large effects sharp structure in $N(\varepsilon)$ can have on the isotope-effect coefficient β . It is not our intention to attempt a realistic estimate of β in Y-Ba-Cu-O. In fact, this system is not likely to be an electron-phonon superconductor.³¹⁻³⁶

Because μ^* was set equal to zero (T_c floats as a and b are changed), the results of Fig. 3 give the effect on β of $N(\varepsilon)$ alone for the several values of a as a function of b. We see that near b = 0, β is near 0.4 from $N(\varepsilon)$ alone, while it peaks at a value near 0.65 before tending toward 0.5 when b gets large. It is clear from this figure that sharp structure in $N(\varepsilon)$ alone can increase or decrease β by a substantial amount over its value for a constant density of states, which is 0.5 when $\mu^*=0$. It substantiates the general trend indicated by the BCS results given in Sec. II. Quantitatively, however, there are serious discrepancies. As an example, it is sufficient to consider



FIG. 3. The coefficient β for the total isotope effect with the Coulomb pseudopotential $\mu(\omega_E)=0$ as a function of position off the Fermi energy b in the Lorentzian form for the electronic density of states $N(\varepsilon)$ given by Eq. (1). The parameters are g = 700 meV, $N_b = 0.5$ and an Einstein spectrum with $\omega_E = 90$ meV is used for the spectral density with mass enhancement parameter $\lambda = 1.5$. Curves are presented for four values of a, namely, $a = 0.25\omega_D$ (solid curve), $a = 0.5\omega_D$ (dotted curve), $a = \omega_D$ (medium-dashed curve), and $a = 2\omega_D$ (long-dashed curve) with $\omega_D = 90$ meV.

the case of b = 0 for which BCS theory gives

$$\beta = \frac{1}{2} \left| 1 - \frac{D}{1+D} \frac{1}{1+\overline{a}^2} \right|$$

and $D = g/(\pi a)$. For the case $a = 0.25\omega_E$ of Fig. 3, $d \approx 2.6$ and $\beta = 0.39$, while BCS theory predicts $\beta \approx 0.3$, which is considerably smaller. This shows clearly that quantitative results can only be obtained through Eliashberg-type calculations.

IV. GENERALIZATION OF THE RAINER-CULETTO FORMULATION

Rainer and Culetto²⁰ have given an elegant formulation of the partial isotope effect $\beta(\omega)$, which applies to phonon modes falling between ω and $\omega + d\omega$ with the total isotope effect given by

$$\beta = \int_0^\infty d\omega \,\beta(\omega) \,\,, \tag{9}$$

with $\beta(\omega) \equiv R(\omega) \alpha^2 F(\omega)$ and

$$R(\omega) = \frac{1}{2} \frac{d}{d\omega} \left[\frac{\omega}{T_c} \frac{\delta T_c}{\delta \alpha^2 F(\omega)} \right].$$
(10)

The weighting function $R(\omega)$ gives the differential isotope effect when multiplied by the value of $\alpha^2 F(\omega)$ at the frequency ω and in that sense can be thought of as an effectiveness function for β .

The functional derivative of T_c with respect to $\alpha^2 F(\omega)$ is well known for an ordinary isotropic superconductor.³⁷⁻³⁹ It is positive definite, exhibiting a single broad maximum around $7k_BT_c$ and goes to zero like ω for $\omega \rightarrow 0$ and like $1/\omega$ for $\omega \rightarrow \infty$. When important energy dependence in the electronic density of states $N(\varepsilon)$ is present, we get modifications. For a peak in $N(\varepsilon)$, $\delta T_c / \delta \alpha^2 F(\omega)$ is significantly depressed over the standard curve for all frequencies, and, at low frequencies, it goes through zero and becomes negative indicating that T_c is now reduced rather than increased by the presence of low-energy phonon modes. Thus, low-energy phonons work like ordinary static impurities, which have the effect of broadening the Lorentzian peak and so reducing the density of states at the Fermi energy, which leads to a drop in T_c .

For a valley in $N(\varepsilon)$, $\delta T_c / \delta \alpha^2 F(\omega)$ closely follows the standard curve at high ω , but near $\omega = 0$ the curve diverges positively like $1/\omega$. This can also be understood in terms of the physical picture that low-frequency phonons act similarly to normal impurities. Their introduction fills in the valley and so increases the effective electronic density of states around the Fermi energy leading to an increase in T_c .

The weighting function $R(\omega)$ for the differential isotope effect $\beta(\omega)$, shown in Fig. 4, very much reflects the shape of the corresponding functional derivative $\delta T_c / \delta \alpha^2 F(\omega)$. It is seen that high-energy phonons are less effective in β than are those at the maximum around $4k_B T_c$. For $\omega \rightarrow 0$, $R(\omega) \rightarrow 0$ in the standard case, but it goes to $-\infty$ like $-1/\omega$ for a peak and to $+\infty$ like $1/\omega$ for a valley. In some cases, at frequencies larger than those shown in Fig. 4, $R(\omega)$ can become negative, and so



 ω/T_{o}

a significant overall negative isotope effect contribution could originate from this region if an important number of phonons falls in that region.

To investigate the partial isotope effect further, we consider a two- δ -function model for the spectral density $\alpha^2 F(\omega)$. The δ functions are centered at 30 and 90 meV, respectively, and each has the same mass renormalization factor $\lambda = 1$ with T_c fixed at 92 K. For the Lorentzian form we take g = 1200 meV with the background $N_{b} = 0.5$. In Fig. 5 (top frame) we show the total isotope effect as a function of b for the same four values of a as in Fig. 3, namely, $a = 0.25\omega_D$ (solid curve), $a = 0.5\omega_D$ (dotted curve), $a = 1\omega_D$ (medium-dashed curve), and $a = 2\omega_D$ (long-dashed curve) with $\omega_D = 90$ meV. What we are really interested in here is a comparison of the total isotope effect β with partial isotope effects β_1 and β_2 coming from a shift of the higher and lower peak (in the spectral density) separately. These are shown, respectively, in the middle and lower frames of Fig. 5. We note that, for small b, β_1 is small and near 0.1 in one case, although the total isotope effect itself is quite large in this region and of the order of 0.3. These small values of β_1 compared with the values for β can be understood from our differential isotope effect results of Fig. 4 where we see that the high-energy phonons have a smaller differential isotope effect $\beta(\omega)$ than the phonons near the maximum in the effectiveness function $R(\omega)$, which falls around $\omega \sim 4k_B T_c$.



0.06

0.04

0.00

-0.02

ල ආ 0.02



FIG. 5. The total isotope effect β (top frame) as a function of b in the Lorentzian form of Eq. (1) compared with the partial coefficient β_1 (middle frame) due to a shift in the $\omega_E = 90$ meV phonons only and β_2 (bottom frame) due to a shift in the $\omega_E = 30$ meV phonons only. The parameters characterizing $N(\varepsilon)$ are g = 1200 meV and $N_B = 0.5$. T_c is 92 K, and the spectral density consists of two peaks, one at 30 meV and the other at 90 meV, each with a mass enhancement factor $\lambda = 1.0$ for a total of 2.0. Four values of a are used, namely, $a = 0.25\omega_D$ (solid curve), $a = 0.5\omega_D$ (dotted curve), $a = \omega_D$ (medium-dashed curve), and $a = 2.0\omega_D$ (long-dashed curve) with $\omega_D = 90$ meV.

Another thing to note, in comparing β_1 and β_2 , is that β_1 shows much more variation with changes in b than does β_2 . While β_1 varies from about 0.1 at b=0 to ~ 0.45 at the maximum for intermediate b and saturates at 0.3 for large b, β_2 varies relatively little around the value of 0.21. A conclusion is that partial isotope effects can be very different from the total β and that energy dependence in $N(\varepsilon)$ can affect each differently, depending on the energy of the phonons involved.

is modified when the energy dependence in the electronic density of states is on the scale of the Debye energy

V. CONCLUSIONS

We have calculated how the isotope effect coefficient β

density of states is on the scale of the Debye energy. While models with fairly freely varying parameters are considered to illustrate the kind of changes we can have, for both total and partial isotope effects, realistic parameters for the case of Nb₃Sn fall in a region of parameter space for which the changes in β over a constant densityof-states approximation are significant. To get further insight into the effect of $N(\varepsilon)$ on β , we also presented results for the differential isotope effect $\beta(\omega)$, which applies to the single-phonon mode ω . To accomplish this, we give a generalization, to include $N(\varepsilon)$, of the formalism of Rainer and Culetto²⁰ developed for a constant electronic density of states. It was found that the effectiveness function $R(\omega)$ for the isotope effect is significantly modified as is the functional derivative of T_c with respect to the electron-phonon spectral density $\alpha^2 F(\omega)$. The changes in $R(\omega)$ are most radical for $\omega \rightarrow 0$. In this limit, for a peak in $N(\varepsilon)$, $R(\omega)$ is found to diverge towards $-\infty$ like $-1/\omega$, while, for a valley, it goes to $+\infty$ like $1/\omega$. This is understood in terms of the effect of low-frequency phonons on T_c , which behave similarly to static impurities. For a peak, T_c decreases because static impurities reduce the density of electronic states, while for a valley T_c increases.

At intermediate frequencies the effectiveness function $R(\omega)$ peaks around $4k_BT_c$ with some variation with material parameters and then drops as ω increases further. At large ω it can be negative and can lead to negative partial and even negative total isotope effects.

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