Role of confined phonons in thin-film superconductivity

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We calculate the critical temperature T_c and the superconducting energy gaps Δ_n of a thin-film superconductor system, where Δ_n is the superconducting energy gap of the nth subband. Since the quantization of both the electron energy and phonon spectrum arises due to dimensional confinement in one direction, the effective electron-electron interaction mediated by the quantized confined phonons is different from that mediated by the bulk phonon, leading to the modification of T_c in the thin-film system. We investigate the dependence of T_c and Δ_n on the film thickness d with this modified interaction.

Superconductivity in thin films has been studied for the last four decades. The phenomenon of thin-film superconductivity has its own specific peculiar features. In early investigations,[1–4] the effect of the film size on the superconducting transition temperature T_c for thin films was investigated. Experiments have shown a monotonic increase of the critical temperature T_c with decreasing film thickness.[4] From the theoretical point of view,[1–3] the shape resonances and the strong thickness dependence of T_c are the characteristic features of thin-film superconductivity. The size quantization of the transverse motion of the electron in the film leads to an increase of T_c with decreasing film thickness, arising essentially from an enhanced effective BCS pairing interaction. The resonance effects are manifest each time one of the n two-dimensional (2D) subband energy levels $E_n(d)$ of a film with thickness d (for 'transverse' motion perpendicular to the film faces or along the confinement direction) passes through the Fermi surface as the thickness d is varied. In previous calculations,[1–3] the phonon modes were assumed to be the same as in the bulk material and only the one-dimensional quantum confinement effects of electrons were considered; i.e., the superconductivity in the thin film was considered to be arising from the attractive electron-electron interaction mediated by bulk phonons interacting with subband-quantized electrons. However, phonons in thin films (''slab'' phonon modes) also have specific characteristic features by virtue of definite boundary conditions imposed by confinement in thin films. The phonon dispersion in a thin film undergoes substantial modification compared with the bulk, and a quantization of the phonon spectrum occurs.[5–7] The quantization of the phonon spectra has many effects on thin-film electronic properties, which have been extensively studied, particularly in the context of semiconductor quantum well structures. It is the purpose of this paper to reconsider the previous calculations[1] of thin-film superconductivity taking into account confined size-quantized phonons in the thin film. We find the resonant shape of the superconducting transition temperature T_c arises from both the quantum electronic confinement and phonon confinement.

Following the BCS theory[8] the critical temperature $T_c \sim 1.14\omega_D \exp(-1/N_0V_0)$ for a bulk BCS superconductor, where ω_D is the phonon Debye frequency, N_0 the electronic density of states at the Fermi energy, and V_0 the effective attractive electron-electron interaction mediated by phonon exchange. An increase of N_0V_0 implies an increase in T_c. For a thin film with thickness d, the density of states at the Fermi energy develops quantum structure due to the confined 2D subbands: $N_{film}=(2\nu-1)m/4\pi d = N_{3D}(2\nu+1)\pi/2dk_F$, where $\nu=1,2,\dots$ is the occupied subband index. Thus when we use the bulk electron-phonon coupling constant J unmodified by any phonon size confinement corrections, which is identical for all subbands, the critical temperature for thin films depends on the film thickness, and decreases exponentially as the thickness increases. However, for a fixed bulk electron density (or, k_F), as the film thickness increases the higher quantized subbands are occupied by Cooper pairs, so that the critical temperature of the film jumps to a higher value due to the higher ν, arising entirely from the jumps in the density of states as the effective Fermi level moves through higher values of the subband index ν. This implies that with increasing thickness of the film the critical temperature of the film exhibits resonance features. In real thin films therefore the electron-phonon coupling constant is different from that in the bulk material due to the quantization of the phonon dispersion,[9] and therefore the simple resonance scenario (discussed above) must be modified. When we consider the confined phonons in the thin film, the electron-phonon coupling constant is different for different subbands, and the effective coupling strength decreases with increasing subband index; that is, higher subbands have progressively weaker electron-phonon coupling constants. This change of the coupling constant gives rise to features in the superconducting energy gap of thin films, which have not earlier been considered in the literature. In this paper we calculate the critical temperature T_c and superconducting energy gaps Δ_n with the modified electron-electron interaction mediated by the confined slab phonons. The results for the calculated critical temperature obviously depend on the slab phonon dispersion, which in turn depends on the boundary conditions used in the phonon confinement model. We have followed here the simple approach of Thompson and Blatt,[1] where the boundary has been treated by an infinite wall, and we use wave functions which vanish at the boundary. Although this is a highly simplified phonon confinement model it has the virtue of being analytically tractable—one could systematically improve upon this model using our theory as the starting point. We expect our results to be valid qualitatively.

We assume that our superconducting film with a finite width d is confined in the z direction by an infinite square-well confinement potential applied at $z=0$ and $z=d$. We choose the same infinite (one-dimensional) square-well confinement for both electrons and phonons, as would be appropriate for a free-standing thin film. Using periodic boundary conditions in the x and y directions with periodicity distance L, we have the one-electron wave function and spectrum

$$\phi_{\mathbf{k},n}(\mathbf{r},z)=u_n(z)\exp(i\mathbf{k}\cdot\mathbf{r})/L,$$

$$\epsilon_n(\mathbf{k})=\frac{\hbar^2k^2}{2m}+E_n. \tag{1}$$

Here, $\mathbf{k}=(k_x,k_y)$, $\mathbf{r}=(x,y)$ are the 2D wave vector and position vector in the plane of free motion, and by solving Schrödinger equation for z direction with confinement potential we have

$$u_n(z)=(2/d)^{1/2}\sin(k_nz),$$

$$E_n=\frac{\hbar^2(k_n)^2}{2m}, \tag{2}$$

where $k_n=n\pi/d$ with $n=$ integer. Thus confinement in the z direction leads to the quantization of electron energy levels into different subbands. In addition to the quantization of electron energy, we take into account the modification in the thin-film phonon dispersion arising from the quantization of the phonon spectrum.[6,9] The quantization of the phonon spectra leads to the change of the conventional electron-phonon interaction. The specific expression for the electron-phonon coupling in the thin film can be obtained on the basis of the general deformation potential electron-phonon interaction theory[10]

$$H_{\text{ep}}=D\int d^3r\Psi^{\dagger}(r)\nabla w(r)\Psi(r), \tag{3}$$

where

$$\Psi(r)=\frac{1}{\sqrt{A}}\sum_{k,n}u_n(z)e^{ik\cdot r}c_{kn}, \tag{4}$$

A is the 2D area, $c_{k,n}$ is the destruction operator for an electron with momentum k in the nth subband, $w(\mathbf{r})$ is the lattice displacement vector, D is the deformation potential coupling constant (which we uncritically assume to be the bulk value since not much experimental information is available). The displacement vector $w(\mathbf{r})$ can be obtained by solving equations of motions of the elastic continuum with the appropriate boundary conditions.[6,9] We skip the details of this elastic continuum theory (which involves substantial algebra) for the sake of brevity.

Within the framework of the BCS theory, Cooper pairs are produced consisting of electrons with opposite $\mathbf{k}$ and identical n in a thin superconducting film, i.e., the electron pairs are produced between $(\mathbf{k},n,\sigma)$ and $(-\mathbf{k},n,-\sigma)$. The direct effective electron-electron interaction due to the exchange of virtual confined slab phonons becomes

$$V_{nm}(q)=\sum_{l=1}^{l_{max}}M_l^{nm}(q)D_l(q)M_l^{mn}, \tag{5}$$

where M_l^{nm} is the electron-confined phonon matrix element and D_l the phonon propagator,[9] and the l sums cover all phonons with energy less than the cutoff energy ω_D. The maximum value of l contributing to the sum over the slab phonon mode l in Eq. (5), l_{max}, is given by the condition $l_{max}=(d/\pi)(\hbar\omega_D/c)$, where c is the velocity of the phonons. (We assume that all slab phonons have the same velocity because information on the slab mode velocity is not experimentally available.) We then get

$$V_{nm}=-\frac{J}{Ad}\sum_{l=1}^{l_{max}}[\beta_{n,n'}^{(l)}]^2, \tag{6}$$

where the confinement form factor is given by

$$\beta_{n,n'}^{(l)}=\frac{2}{d}\int_0^d dz u_n^*(z)\sin(l\pi z/d)u_{n'}(z)$$

$$=\frac{2}{\pi}\left[\frac{l}{l^2-(n-n')^2}-\frac{l}{l^2-(n+n')^2}\right]. \tag{7}$$

Equation (7) has the following selection rule: for $|n\pm n'|$ = even (odd) only odd (even) l's are allowed in the sum over slab modes l. The slab phonons with odd l are symmetric and the ones with even l are antisymmetric with respect to reflection through $z=d/2$, i.e., under the transformation $|z-d/2|\to|d/2-z|$. Since the electron wave functions are either symmetric or antisymmetric, the couplings between two subbands of the same symmetry involve symmetric quantized phonons, while couplings between two subbands of different symmetry involve antisymmetric phonons. (Note that this simplicity will be lost if parity is not a good quantum number as it is in our simple infinite square-well model, but would not be under an asymmetric confinement.) We see that V_{nm} decreases with increasing l, since the transition of electrons to higher subbands cannot be induced by phonons with small momenta q. Thus the components V_{nm} of the interaction matrix form a monotonically decreasing sequence with increasing subband index. When all the confined phonons contribute to V_{nm} (i.e., $l_{max}\to\infty$) we recover the bulk phonon mediated results[1] i.e., $V_{nm}=-(J/Ad)(1+\frac{1}{2}\delta_{nn'})$, as we should. As a result of the confined phonons, a superconducting condensate of Cooper pairs can be produced in a given subband, first because of the attraction due to the electrons in the same subband and second because of transitions from other subbands contributing to the condensate.

In the presence of a number of subbands the reduced BCS Hamiltonian of the system is given by

$$H=\sum_{\mathbf{k},n,\sigma}\xi_n(k)c_{\mathbf{k}n\sigma}^{\dagger}c_{\mathbf{k}n\sigma}$$

$$+\sum_{\mathbf{k},\mathbf{k}',\sigma}\sum_{n,m}V_{nm}c_{\mathbf{k}'m\sigma}^{\dagger}c_{-\mathbf{k}'m-\sigma}^{\dagger}c_{\mathbf{k}n\sigma}c_{-\mathbf{k}n-\sigma}, \tag{8}$$

where $c^{\dagger}_{\mathbf{k}n\sigma}$ is electron creation operator in the nth subband with spin σ, $\xi_n(k)=\epsilon_n(k)-\mu$ the electron energy in the nth subband measured from the chemical potential μ, and V_{nm} the attractive interaction between nth subband and mth subband mediated by the confined phonons in the film.

In the BCS theory (which is what we utilize) the gap function has the same energy cutoff $\hbar\omega_D$ as the interaction. In the weak-coupling approximation we have the the superconducting energy gap for nth subband given by the gap equation

$$\Delta_n(T)=\sum_{m\mathbf{k}}\frac{V_{nm}}{2V}\frac{\Delta_m\tanh(E_m/2kT)}{E_m}, \tag{9}$$

where $E_n=(\xi_n^2+\Delta_n^2)^{1/2}$. The nondiagonal terms in the sum reflect the possibility of the transition of the electron pair from one subband into another as a result of interaction with confined phonons. Integration over k gives the gap function of the subband n at $T=0$ K:

$$\Delta_n=\frac{Jm}{2\pi}\sum_{n'}\sinh^{-1}\left(\frac{\omega_c}{\Delta_{n'}}\right)\Delta_{n'}\alpha_{nn'}, \tag{10}$$

where $\alpha_{nn'}=V_{nn'}/J$. If all confined phonons contribute equally to the electron-electron interaction, i.e., ($\alpha_{nm}=1+\delta_{nn'}/2$), then we recover the results of Ref. 1. With the coupled subband interactions Eq. (10) becomes a nonlinear coupled subband matrix equation. The critical temperature is given by

$$T_c=1.14\omega_D\exp\left(-\frac{2\pi d}{Jm\sum_{n'}\alpha_{1n'}x_{1n'}}\right), \tag{11}$$

where $x_{1n}=\Delta_n/\Delta_1$, the ratio of the nth subband energy gap to the lowest subband energy gap. For any given finite width d of the slab, only a finite number of eigenvalues ξ_n contribute; values of ξ_n in excess of $\mu+\hbar\omega_D$ make vanishing contributions, because then all the $\varepsilon_{\mathbf{k}}$'s lie outside the interaction region. Thus the summation in Eqs. (10) and (11) is only over all the occupied subbands. For a fixed electron density we can find the maximum value of the n (or, the highest occupied subband) from the chemical potential. The number density $n_0=N/V$ is related to the chemical potential by the relation, $N=2\Sigma_{\mathbf{k},n}n_{\mathbf{k},n}$, where $n_{\mathbf{k},n}=[\exp(-\varepsilon_n(\mathbf{k})/k_BT)+1]^{-1}$ is the Fermi distribution function. At $T=0$ K we have

$$\mu=(\pi d\hbar^2/\nu M)\left\{n_0+\frac{\pi}{6d^3}\nu\left(\nu+\frac{1}{2}\right)(\nu+1)\right\}, \tag{12}$$

where ν is the maximum value of occupied subband n and is given by the integral value of the expression, $\nu=dk_F/\pi$. If we write Eq. (11) in the usual BCS form, $T_c=1.14\omega_D\exp(-1/N_{film}J_{eff})$, the effective interaction parameter can be written as

$$J_{eff}=\frac{2J}{2\nu+1}\sum_{n=1}^{\nu}\alpha_{1n}x_{1n}. \tag{13}$$

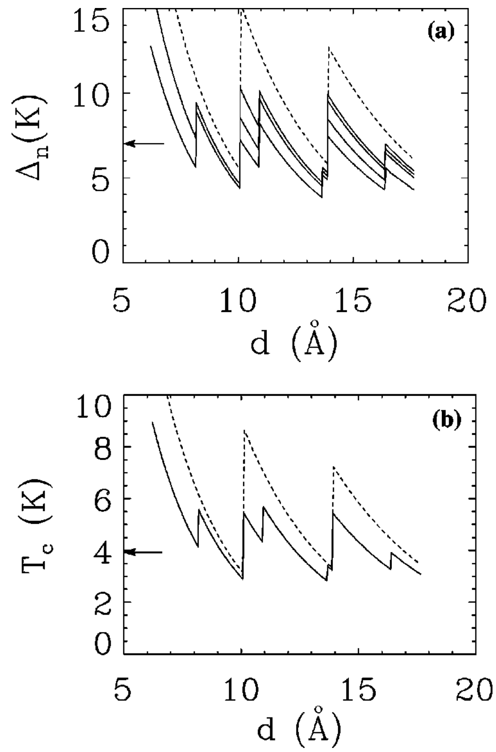

FIG. 1. (a) The calculated superconducting energy gaps Δ_n of each occupied subband n, and (b) the critical temperature for an electron density $n=2\times10^{22}$ cm^{-3} as a function of the film thickness d. In (a) the highest curve is the energy gap of the lowest subband ($n=1$), and the second highest curve is that of the first excited subband ($n=2$), and so on, with four occupied subbands. In (b) the solid line indicates our result with the interaction mediated by confined slab phonons and the dashed line from Ref. 1 corresponds to the bulk phonon result. The arrow in each figure indicates the purely bulk result.

As $d\rightarrow\infty$, $J_{eff}\rightarrow J$ because $\alpha_{1n}(d\rightarrow\infty)=1+\delta_{1n}/2$ and $x_{1n}(d\rightarrow\infty)=1$, and $N_{film}\rightarrow N_{3D}$. Thus, we recover the three-dimensional results as we expect.

In Fig. 1 we show the calculated superconducting energy gaps of each subband and the critical temperature as a function of the film thickness d up to the fourth subband occupation. In this figure we use the following parameters: Debye energy $\hbar\omega_D=100$ K, electron density $n_0=2\times10^{22}$ cm^{-3}, and $\rho=N_{3D}J=mk_FJ/(2\pi^2)=0.3$. In previous calculations[1,2] the energy gaps of different subbands were find to be the same by virtue of the bulk phonon approximation. The shape resonance feature in the earlier calculations[1,2] arise only from the effective 2D density of state of the film as the chemical potential passes through different subbands. In our calculation we find that the energy gaps are different for different subbands since the effective coupling strength depends explicitly on the occupied subbands. In addition, the resonance structures in our results (the sharp maxima in Fig. 1) arise from both the thickness dependent density of states N_{film} and the effective interaction parameter J_{eff}. The energy gap is maximum for the lowest subband and decreases in the higher subbands for a fixed film thickness. In Fig. 1(b) we also compare our results with Ref. 1 for equivalent parameter values. Our results exhibit more resonance features and has much lower T_c (typically, about half around the maxima) than that of Ref. 1. Note that any inhomogeneity on the microscopic scale (quite unavoidable in real thin films of 5–15-Å thickness) will considerably suppress the resonance features of Fig. 1, and any enhancement in Δ_n or T_c may remain unobservable unless the films are microscopically of uniform thickness. The reduction of the critical temperature in our calculation compared with that obtained in Ref. 1 [note that our calculated T_c is still enhanced above the bulk T_c as is obvious from Fig. 1(b)] can be explained by the enhancement of the effective interaction parameter in the slab phonon model.

Our main approximation is that we use an infinite surface barrier for both electrons and phonons. The present work can easily be extended to include more realistic boundary conditions,[2] but one then needs to resort to numerical work right from the beginning, losing much of the essential qualitative physics of the phenomenon. The infinite barrier approximation is obviously only of qualitative validity, but in general this approximation usually works well for phonons unless the dispersion curves for adjacent regions overlap. In our calculation we treat the boundaries as impenetrable in order to simplify the calculations. In real systems the boundaries become softer at the surface, so that electrons inside the slab may interact with surface phonons. In particular, the surface phonons may induce electrons to form Cooper pairs at the boundaries and give rise to increase in T_c.[11] Thus in our strict ''infinite wall'' model the enhancement of T_c is smaller than that in the more realistic boundary conditions. Our calculated T_c could therefore be considered to be lower bound for the expected T_c's in thin-film superconductors within the BCS model. We believe that the basic physics discussed in this paper and the qualitative features of our results shown in Fig. 1 transcend our specific model, and should be valid in any BCS-type superconductivity in thin films.

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