Boundary condition for Ginzburg-Landau theory of superconducting layers

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(Received 27 January 2009; published 7 May 2009)

Electrostatic charging changes the critical temperature of superconducting thin layers. To understand the basic mechanism, it is possible to use the Ginzburg-Landau theory with the boundary condition derived by de Gennes from the BCS theory. Here we show that a similar boundary condition can be obtained from the principle of minimum free energy. We compare the two boundary conditions and use the Budd-Vannimenus theorem as a test of approximations.

DOI: 10.1103/PhysRevB.79.174510 PACS number(s): 74.20.De

I. INTRODUCTION

Much experimental effort is devoted to find superconducting materials with critical temperatures as high as possible. It is well known that the critical temperature depends on the charge-carrier density. The charge-carrier density can be changed by doping, and to some extend it can also be changed by electrostatic charging. Consequently it is an attractive task to determine, how electrostatic charging evoked by an applied electric field changes the critical temperature of superconductors. Generally the experiments revealed that it is easier to increase T_c than to decrease it.[1,2] The Ginzburg-Landau (GL) theory with the de Gennes boundary condition can be used to understand this behavior.[3]

The field effect on the superconductivity can be sketched as a two-step process. First, the field polarizes the surface pulling electrons out or pushing them in. This results in a change in the electrostatic potential which modifies the local density of states. Second, the electrostatic potential affects the superconductivity via the density of states. The second step has been studied within various approximations ranging from the GL theory[4] over the Bogoliubov–de Gennes method[5] and the de Gennes boundary condition[3] up to Nambu-Gor'kov Green's functions.[6–8] In these studies the electrons of a metal were restricted to a half space, and their response to the applied field was described by a simple exponential screening.

The aim of this paper is to describe the interaction of the field with electrons in more detail. We show how a superconductor screens the external electric field and to which extent the boundary condition derived from the minimum free-energy principle is compatible with the de Gennes boundary condition.

Charges at a solid surface partially leak out of the surface. This creates a surface dipole. The Budd-Vannimenus theorem[9] describes the step in the surface potential due to this surface dipole as a simple expression of the bulk free-energy density. Therefore it is well suited to test the approximations used in this paper.

In Sec. I we explain the model and the parts considered in the free energy of the superconductor and solve the Euler-Lagrange equations for the GL and charge-carrier wave function and the surface potential. In Sec. III the corresponding equations outside the superconductor are solved, and in Sec. IV the continuity requirements determine the remaining constants. Section V presents the numerical values which are compared with the de Gennes boundary condition in Sec. VI. Finally we conclude in Sec. VII.

II. FREE ENERGY IN THE SUPERCONDUCTOR

We start with the free energy

$$\mathcal{F}=\int (f_{\mathrm{TF}}+f_{\mathrm{GC}}+f_{\mathrm{GL}}+f_{\mathrm{elst}})d\mathbf{r}, \tag{1}$$

where we include only the terms most relevant for the above specified problem.

The first term f_{TF} is the Thomas-Fermi internal energy, for which we use the local-density approximation (LDA),

$$f_{\mathrm{TF}}=\frac{3}{5}(3\pi^2)^{2/3}\frac{\hbar^2}{2m}n^{5/3}. \tag{2}$$

The second term f_{GC} represents the condensation energy, for which we use the formula following from the Gorter-Casimir two fluid model,[10,11]

$$f_{\mathrm{GC}}=\frac{1}{4}\gamma T_c^2\left(\frac{n_s}{n}+2\frac{T^2}{T_c^2}\sqrt{1-\frac{n_s}{n}}\right). \tag{3}$$

The electrostatic energy density term reads

$$f_{\mathrm{elst}}=-\frac{1}{2}\epsilon_0\mathbf{E}^2+e\varphi\delta n, \tag{4}$$

in the form suitable for performing variations. For simplicity we exclude the magnetic field and its related kinetic energy of the screening current. We take the vector potential $\mathbf{A}$ to be zero and write the GL gradient term as

$$f_{\mathrm{GL}}=\frac{\hbar^2}{2m^*}\psi_n^2|\nabla\psi|^2. \tag{5}$$

Here we have chosen the GL wave function ψ normalized with respect to the total charge-carrier density n. In the spirit

of the Thomas-Fermi approximation the charge carriers are described by a wave function ψ_n with $n=\psi_n^2$ and the superconducting fluid density used in the formula for the condensation energy [Eq. (3)] reads

$$n_s = 2\psi_n^2\psi^2. \tag{6}$$

In short, the free energy is expressed by three independent variables: the scalar potential φ determining the electric field $\mathbf{E}=-\nabla\varphi$, the GL wave function ψ, and the charge-carrier wave function ψ_n. We assume that the material parameters, the critical temperature T_c, and the Sommerfeld parameter γ depend on the charge carrier's density n, by using the following approximations:

$$\gamma(n) = \gamma_0\left(1 + \frac{n-n_{\rm lat}}{n_{\rm lat}}\frac{\partial \ln \gamma}{\partial \ln n}\right), \tag{7}$$

$$T_c(n) = T_{c0}\left(1 + \frac{n-n_{\rm lat}}{n_{\rm lat}}\frac{\partial \ln T_c}{\partial \ln n}\right), \tag{8}$$

where $n_{\rm lat}$ is the crystal lattice density. In the following we shall use three characteristic lengths: (i) the Thomas-Fermi screening length $\lambda_{\rm TF}^2=\frac{2\epsilon_0}{3ne^2}E_F$, (ii) the Bohr radius $a_B=\frac{4\pi\epsilon_0\hbar^2}{me^2}$, and (iii) the coherence length $\xi_0^2=\frac{\hbar^2 n_{\rm lat}}{4\gamma_0 T_{c0}^2 m}$.

From the charge neutrality requirement we know that $\psi_{n\infty}=\sqrt{n_{\rm lat}}$ (here and in the following the subscript ∞ denotes the magnitude far from the surface). To keep things simple, we use the following approximations:

$$\psi_n = \sqrt{n_{\rm lat}}(1+\delta\psi_n), \tag{9}$$

$$\psi = \psi_\infty(1+\delta\psi), \tag{10}$$

$$e\varphi = E_F(\varphi_\infty + \delta\varphi), \tag{11}$$

and suppose that the deviations of the three independent variables from the optimum values are small. In a homogeneous superconductor far from the surface all these deviations are zero and the derivatives of the free-energy formula [Eq. (1)] with respect to them must be also zero. From these requirements we get the magnitudes of the optimum superfluid fraction

$$\frac{\psi_\infty}{\psi_{n\infty}} = \sqrt{1-t^4} \tag{12}$$

and the optimum magnitude of the scalar potential

$$\varphi_\infty = -1 + \frac{2\lambda_{\rm TF}^4}{\pi^2 a_B^2 \xi_0^2}\left[2(1-t^4)\frac{\partial \ln T_c}{\partial \ln n} + (1+t^4)\frac{\partial \ln \gamma}{\partial \ln n}\right]. \tag{13}$$

The electrostatic potential energy of the charge carrier thus equals the Fermi energy

$$E_F = \frac{\hbar^2}{2m}(3\pi^2 n)^{2/3}, \tag{14}$$

with a small (lower than gap) correction represented by the second term in Eq. (13).

Using the second-order expansion of the free energy [Eq. (1)], from the variation we get three linear Euler-Lagrange (EL) equations for the three independent variables, $\delta\psi_n$, $\delta\psi$, and $\delta\varphi$ as

$$\frac{3}{4}\lambda_{\rm TF}^2\nabla^2\delta\varphi + \delta\psi_n = 0, \tag{15}$$

$$(1-t^4)\xi_t^2\nabla^2\delta\psi + t^4\delta\psi + 4t^4\frac{\partial \ln T_c}{\partial \ln n}\delta\psi = 0, \tag{16}$$

$$\left[2t^4\frac{\partial \ln T_c}{\partial \ln n}\left(\frac{\partial \ln T_c}{\partial \ln n} + 2\frac{\partial \ln \gamma}{\partial \ln n}\right) - \frac{\pi^2 a_B^2 \xi_t^2}{12\lambda_{\rm TF}^4}\right]\delta\psi_n + 2t^4\frac{\partial \ln T_c}{\partial \ln n}\delta\psi - \frac{\pi^2 a_B^2 \xi_t^2}{16\lambda_{\rm TF}^4}\delta\varphi = 0. \tag{17}$$

Close to the planar surface we can assume exponential dependencies of the deviations and from the EL Eqs. (15)–(17), we get a second-order equation for the square of the expected penetration depth λ. Two solutions arise out of it.

Observing that $\lambda_{\rm TF}$, $a_B \ll \xi_0$ we find a first approximate solution λ in the form of the coherencelike length

$$\xi_t = \xi_0\frac{2t^2}{\sqrt{1-t^4}}. \tag{18}$$

In this solution the scalar potential is constant and local charge neutrality is preserved. Only the deviation of the wave function $\delta\psi$ is nonzero (C_ξ will denote its magnitude).

A very small penetration depth characterizes the second solution such that this solution can be simplified. Using the same approximation as above, we find that the second penetration depth equals the Thomas-Fermi screening length $\lambda_{\rm TF}$. In this solution the scalar potential displays a sharp step ($C_{\rm TF}$ will denote its magnitude) and from the Poisson Eq. (15) follows that the charge carrier's density changes accordingly. The sharp step on the GL wave function ψ is negligibly small due to the factor $\lambda_{\rm TF}^2/\xi_t^2$ which enters the resulting formula. It corresponds to the well-known fact that the GL wave function ψ cannot abruptly change.

The general solution can be written as a sum of the two above-described solutions:

$$\delta\varphi = C_{\rm TF}\exp\left(\frac{-x}{\lambda_{\rm TF}}\right), \tag{19}$$

$$\delta\psi_n = -\frac{3}{4}C_{\rm TF}\exp\left(\frac{-x}{\lambda_{\rm TF}}\right), \tag{20}$$

$$\delta\psi = \frac{3\lambda_{\rm TF}^2 t^4}{(1-t^4)\xi_t^2}\frac{\partial \ln T_c}{\partial \ln n}C_{\rm TF}\exp\left(\frac{-x}{\lambda_{\rm TF}}\right) + C_\xi\exp\left(\frac{-x}{\xi_t}\right). \tag{21}$$

Here $C_{\rm TF}$ describes the step of the scalar potential in units of E_F/e according to Eq. (11). Using Eq. (19)–(21) we can calculate the free energy

$$\mathcal{F}=\int\left(\frac{3}{5}+\frac{2(1+t^4)\lambda_{\mathrm{TF}}^4}{\pi^2 a_B^2\xi_0^2}\right)d\mathbf{r}-\frac{3}{4}\lambda_{\mathrm{TF}}C_{\mathrm{TF}}^2+\frac{8\lambda_{\mathrm{TF}}^4(1-t^4)}{\pi a_B^2\xi_t}C_\xi^2+\frac{48t^4\lambda_{\mathrm{TF}}^5}{\pi^2 a_B^2\xi_t^2}\frac{\partial \ln T_c}{\partial \ln n}C_{\mathrm{TF}}C_\xi. \tag{22}$$

For a semi-infinite medium the first term gives an infinite contribution which is not influenced by the surface conditions, so we do not need to deal with it. The last three terms correspond to the surface energy, which according to the principle of minimum free energy should take an extremum. The minimum of the free energy is obtained for

$$C_\xi=-\frac{3\lambda_{\mathrm{TF}}\xi_t}{4\xi_0^2}\frac{\partial \ln T_c}{\partial \ln n}C_{\mathrm{TF}}, \tag{23}$$

in which case the derivative of the wave function ψ at the surface is zero. For lead at $t=0.9$ we get $C_\xi=-0.000\,44C_{\mathrm{TF}}$. As expected, the deviation of the GL wave function $\delta\psi$ is much smaller compared to the sharp steps on the scalar potential $\delta\varphi$ and on the charge-carrier wave function $\delta\psi_n$.

We see that the principle of minimum free energy entails the GL boundary condition. Toward the surface the GL wave function $\delta\psi$ displays a small gradual change, only very close to the surface its derivative jumps to zero. The solution is complete if the parameter C_{TF} is determined. It can be derived from the requirement of continuity with a solution minimizing the total free energy including the one of the vacuum outside.

III. FREE ENERGY OUTSIDE THE SLAB

Now we approximate the free-energy density outside the superconductor by

$$\mathcal{F}=\int(f_W+f_{\mathrm{GL}}+f_{\mathrm{elst}})d\mathbf{r}. \tag{24}$$

We include the electrostatic term, the GL gradient correction, and the von Weizsäcker kinetic-energy functional,

$$f_W=\frac{\hbar^2}{2m}|\nabla\psi_n|^2. \tag{25}$$

In the limit of rapidly varying densities this kinetic-energy term is dominant, and when describing charge carriers tunneling outside the superconductor this term cannot be omitted. We have not included this term into formula (5) describing the free energy inside. The reason is that inside the superconductor the Thomas-Fermi internal energy plays the dominant role and moreover, as it is shown, e.g., in the book of Dreizler and Gross,[12] in the limit of nearly homogeneous systems the second-order term of the gradient expansion provides a better approximation. It has the same structure as the von Weizsäcker kinetic-energy functional, but its coefficient is nine times lower. We suppose that for the rough estimates presented here this relatively small correction can be neglected.

In the vacuum far from the surface the scalar potential reaches the magnitude of the work function φ_W so that we can approximate

$$e\varphi=E_F(\varphi_W+\delta\varphi). \tag{26}$$

The density of the tunneling charge carriers quickly drops to zero. Using an analogous notation as above we write

$$\psi_n=\sqrt{n_{\mathrm{lat}}}\delta\psi_n, \tag{27}$$

supposedly that $\delta\psi_n$ is small. For the wave function ψ we use the approximation

$$\psi=\tilde{\psi}_\infty+\delta\psi, \tag{28}$$

where $\tilde{\psi}_\infty$ represents the superfluid fraction in the vacuum far from the surface. Let us remind that ψ is normalized to the charge-carrier density, see Eq. (6), so that $\tilde{\psi}_\infty$ does not need to be zero. The free-energy density in the vacuum thus reads

$$f_{\mathrm{out}}=\frac{8\lambda_{\mathrm{TF}}^4}{\pi^2 a_B^2}[2(\nabla\delta\psi_n)^2+\delta\psi_n^2(\nabla\psi)^2]-\frac{3}{4}\lambda_{\mathrm{TF}}^2(\nabla\delta\varphi)^2+(\varphi_W+\delta\varphi)\delta\psi_n^2, \tag{29}$$

and we can write the Euler-Lagrange equations.

The variation with respect to the wave function ψ gives the condition

$$\delta\psi_n^2\nabla^2\delta\psi=0. \tag{30}$$

We see that $\delta\psi$ remains constant or changes linearly.

The proximity effects indicate that the correlated charge carriers can remain correlated even if they are tunneling. For simplicity we suppose that the superfluid fraction of the charge carriers tunneling outside the material does not change, so we take $\delta\psi=\psi(0)$. The two other Euler-Lagrange equations read

$$2\varphi_W\delta\psi_n-\frac{32\lambda_{\mathrm{TF}}^4}{\pi^2 a_B^2}\nabla^2\delta\psi_n=0, \tag{31}$$

$$\delta\psi_n^2+\frac{3}{2}\lambda_{\mathrm{TF}}^2\nabla^2\delta\varphi=0. \tag{32}$$

In the same way as above we can try the exponential solution

$$\delta\psi_n=K_n\exp\left(\frac{x}{\lambda_W}\right), \tag{33}$$

$$\delta\varphi=K_\varphi\exp\left(\frac{2x}{\lambda_W}\right), \tag{34}$$

where λ_W denotes the tunneling length which follows from the Euler-Lagrange Eq. (32) as

$$\lambda_W=\frac{\sqrt{-6K_\varphi}}{K_n}. \tag{35}$$

The work function can be determined from Eq. (31) as

$$\varphi_W=\frac{16\lambda_{\mathrm{TF}}^4}{\pi^2 a_B^2\lambda_W^2}. \tag{36}$$

In this way we have an approximate solution outside the superconductor, which should be linked to the solution inside.

IV. CONTINUITY REQUIREMENTS

At the surface the continuity of the wave function ψ_n and the continuity of the scalar potential φ with its derivative (continuity of the electric field) must be ensured. We get three conditions,

$$\frac{2\lambda_{\mathrm{TF}}^4}{\pi^2 a_B^2 \xi_0^2}\left[2(1-t^4)\frac{\partial \ln T_c}{\partial \ln n}+(1+t^4)\frac{\partial \ln \gamma}{\partial \ln n}\right]-1+C_{\mathrm{TF}} = K_\varphi - \frac{8\lambda_{\mathrm{TF}}^2}{3\pi^2 a_B^2}\frac{K_n^2}{K_\varphi}, \tag{37}$$

$$-C_{\mathrm{TF}} = \frac{2K_\varphi K_n}{\sqrt{-6K_\varphi}} + E_a, \tag{38}$$

$$1-\frac{3}{4}C_{\mathrm{TF}} = K_n \tag{39}$$

where the term E_a representing the applied electric field is included into the condition of continuity for the electric field. From the continuity requirements Eqs. (37)–(39) we obtain the equation

$$\frac{2\lambda_{\mathrm{TF}}^4}{\pi^2 a_B^2 \xi_0^2}\left(2(1-t^4)\frac{\partial \ln T_c}{\partial \ln n}+(1+t^4)\frac{\partial \ln \gamma}{\partial \ln n}\right) - \frac{\lambda_{\mathrm{TF}}^2(-4+3C_{\mathrm{TF}})^4}{144\pi^2 a_B^2 (C_{\mathrm{TF}}-E_a)^2} + \frac{24(C_{\mathrm{TF}}-E_a)^2}{(-4+3C_{\mathrm{TF}})^2} - 1 + C_{\mathrm{TF}} = 0, \tag{40}$$

determining the step of the scalar potential C_{TF}.

V. NUMERICAL VALUES

The sixth-order Eq. (40) can be numerically solved. For small applied electric fields the linear expansion

$$C_{\mathrm{TF}} = C_{\mathrm{TF0}} + \zeta E_a \tag{41}$$

is applicable, and for lead at temperature $t=0.9$ we get the numerical solution

$$C_{\mathrm{TF}} = 0.457 - 0.53 E_a. \tag{42}$$

The numerical estimate for the tunneling length follows from Eq. (35) to be $\lambda_W = 3.17\lambda_{\mathrm{TF}}$ and the work function according to Eq. (36) of $\varphi_W = 1.43$ eV. Taking into account how many simplifications we have used, it is surprising that the obtained results seem to be quite reasonable. The estimated magnitude of the work function is comparable with the experimentally determined value of $\varphi_W = 4.25$ eV.[13]

The sharp step of the scalar potential can be estimated from the modified Budd-Vannimenus theorem[9] according to which

$$e(\varphi_\infty - \varphi_0) = \left(\frac{\partial f_{\mathrm{el}}}{\partial n} - \frac{f_{\mathrm{el}}}{n}\right). \tag{43}$$

Here f_{el} denotes the spatial density of the electronic free energy, which can be roughly approximated by the Thomas-Fermi internal energy f_{TF} defined in Eq. (2). Then the Budd-Vannimenus theorem [Eq. (43)] predicts a sharp step, $C_{\mathrm{TF}} = \frac{2}{5}$, of the scalar potential in units of E_F/e. The numerical solution [Eq. (42)] gives a comparable result what strongly supports the applicability of the here used approximations.

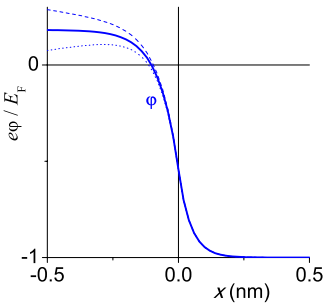

FIG. 1. (Color online) The electrostatic potential φ calculated for lead at $t=0.9$ K. The dashed and dotted lines correspond to applied electric field $E_a = \pm 0.01$.

We saw that the numerical values of the measurable quantities are reasonable. In Fig. 1 the scalar potential is plotted. As expected, inside the superconductor the scalar potential acquires the Fermi energy value, while in the vacuum outside it reaches the work function value φ_W. The dashed and dotted lines correspond to the experimentally accessible applied electric field $E_a = \pm 0.01 \frac{E_F}{e\lambda_{\mathrm{TF}}} \cong \pm 1.7 \times 10^7$ V/cm. As it is seen in Fig. 2, the external electric field is screened on the Thomas-Fermi screening length. In the Fig. 3 deviations of the charge-carrier densities from the equilibrium values are plotted. We can see that the magnitudes of these deviations are small. Close to the surface the superfluid density n_s decreases, and this decrease is compensated by an increase in the normal fluid density n_n. The total charge-carrier density n shows no change on the scale of the coherence length.

VI. COMPARISONS WITH DE GENNES FORMULA

Now we compare the GL boundary condition following from the minimum free-energy principle with the de Gennes boundary condition[14]

$$\left.\frac{\nabla\psi}{\psi}\right|_0 = \left.\frac{\nabla\Delta}{\Delta}\right|_0 = \frac{1}{b} = \frac{1}{b_0} + \frac{E_a}{U_s}, \tag{44}$$

according to which the derivative of the gap at the surface is not exactly zero even without external electric field. The

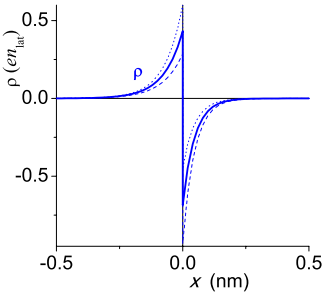

FIG. 2. (Color online) Charge density ρ. To make the screening visible, the dashed and dotted lines correspond to the applied electric field $E_a = \pm 0.3$.

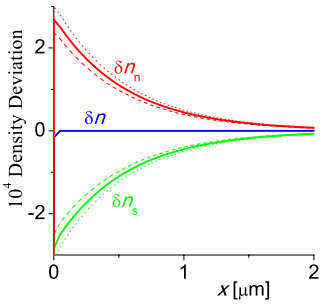

FIG. 3. (Color online) Deviations of charge-carrier densities. To ensure visibility, the dashed and dotted lines correspond to the applied field $E_a = \pm 0.1$.

zero-field extrapolation length b_0 is around 1 cm (almost infinity from the microscopic point of view). The effective potential U_s,

$$\frac{1}{U_s} = \frac{3\eta\lambda_{\mathrm{TF}}^2}{2\xi_0^2}\frac{\partial \ln T_c}{\partial \ln n}\frac{e}{E_F}, \tag{45}$$

determines how the extrapolation length b changes if an external electric field E_a is applied.[3] De Gennes estimated the surface ratio,

$$\eta \equiv \frac{\Delta(0)}{\Delta_0}, \tag{46}$$

to be close to one. For lead formula (45) gives $U_s = 1.35 \times 10^7$ V. From the minimum free energy we know, however, that the derivative at the surface should be zero. In Fig. 4 we see how the deviation of the wave function ψ at the surface decreases with the derivative determined by the parameter C_ξ. From this we get the extrapolation length $b_0 \approx 2.8$ mm, a value comparable with the one estimated by de Gennes. Only very close to the surface (on the distance of the Thomas-Fermi screening length) the derivative of the wave function ψ approaches zero (see insert of the Fig. 4).

In Fig. 4 we can also observe how the extrapolation length changes if an electric field E_a is applied. By substituting $b = -\xi_t / C_\xi$ into Eq. (44) and using Eq. (23) with the approximation [Eq. (41)], we get a simple expression for the effective potential U_s,

$$\frac{1}{U_s} = \frac{3\zeta\lambda_{\mathrm{TF}}^2}{4\xi_0^2}\frac{\partial \ln T_c}{\partial \ln n}\frac{e}{E_F}. \tag{47}$$

This formula is similar to the de Gennes formula [Eq. (45)]. We should notice, however, that in this formula the extrapolation parameter $\zeta/2$ of Eq. (41) appears instead of the surface ratio η which enters the de Gennes formula [Eq. (45)].

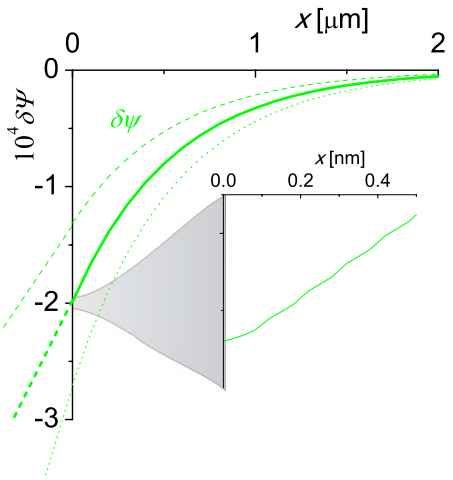

FIG. 4. (Color online) Deviation $\delta\psi$ of the wave function ψ from the equilibrium value. Close to the surface its derivative approaches zero (see the insert). The dashed and dotted lines indicate how the extrapolation length changes with applied field.

VII. CONCLUSIONS

It was shown in this paper that the minimum free-energy principle entails a zero derivative of the wave function ψ at the surface of the superconductor. On the scale of the coherence length, however, even if no external electric field is applied, the derivative is nonzero and its magnitude corresponds to the de Gennes estimate. Only on the Thomas-Fermi screening length scale it approaches zero. In the presence of an external electric field the extrapolation length changes according to Eq. (44), with the effective potential given by Eq. (47). This formula is similar to formula (45) following from the de Gennes theory. The agreement with the Budd-Vannimenus theorem and the numerical estimates support the applicability of the proposed approach.

The present method does not include the periodic potential of ionic background. This jelly model is applicable to simple metals, but it is not justified for systems with strong space modulations of the electron charge, e.g., layered structures of high-T_c materials.

ACKNOWLEDGMENTS

This work was supported by the Czech research plans MSM under Grants No. 0021620834 and No. AVOZ10100521, by grants GAČR under Grants No. 202/07/0597 and 202/08/0326 and GAAV under Grant No. IAA100100712 as well as German PPP project of DAAD and the BMBF. The financial support by the Brazilian Ministry of Science and Technology is acknowledged.

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