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NOVEL RESULTS ABOUT MAGNETIC FLUCTUATION EFFECTS NEAR THE NORMAL-TO-SUPERCONDUCTING PHASE TRANSITION IN A ZERO MAGNETIC FIELD

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A systematic treatment of the magnetic fluctuations effect on the properties of the normal-to-superconducting phase transition in a zero external magnetic field is given within the self-consistent approximation and the quasi-macroscopic Ginzburg–Landau model. New results for thin superconducting films are presented. Thermodynamic quantities having a direct experimental interest as the order parameter jump, latent heat, and specific heat are considered and numerically evaluated for bulk Al and thin Al films. The possibility for an experimental verification of the theoretical predictions is discussed.

Key words: latent heat, order parameter, phase transition, equation of state.

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I. INTRODUCTION

In 1974 Halperin, Lubensky and Ma (HLM) [1] showed that the magnetic fluctuations change the order of the superconducting phase transition in a zero external magnetic field ($H_0 = |\mathbf{H}_0| = 0$)), i.e., the order of the phase transition from normal-to-uniform (Meissner) superconducting state at $T_{c0} = T_c(H_0 = 0)$. In the mean-field approximation, when both magnetic and superconducting fluctuations are neglected, this phase transition is of second order; see, e.g., Refs. [2,3]. Moreover, the fluctuations $\delta\psi(\mathbf{x}) = [\psi(\mathbf{x}) - \langle\psi(\mathbf{x})\rangle]$ of the superconducting order parameter $\psi(\mathbf{x})$ towards the statistical average $\langle\psi(\mathbf{x})\rangle$ are extremely small and can be safely ignored in usual low-temperature ($T_{c0} < 20$ K) superconductors. For a long time these superconductors have been considered as an excellent example of a standard phase transition of second order described by the mean-field approximation. When the magnetic fluctuations are taken into account in the Ginzburg–Landau (GL) free energy $F(\psi, \mathbf{A})$ of superconductor [2], the same normal-to-superconducting phase transition in a zero (mean) external magnetic field ($\mathbf{H}_0 = 0$) is found to be a weakly-first order phase transition with a very small latent heat which cannot be observed by available experimental techniques [1]. The effect of a magnetic fluctuation change of the superconducting phase transition order, called HLM effect, is very weak in bulk (three dimensional, or 3D) superconductors even in Al where the GL number κ is very small ($\kappa \ll 1$) — a circumstance which is in favor of the effect [1,4].

In this paper we shall investigate this fluctuation-induced first order phase transition in thin (quasi-2D) superconducting films. Bulk superconductors will be also discussed in order to compare them with the behaviour of thin films. We shall use a self-consistent approximation [1], in which the fluctuations $\delta\psi$ of ψ are neglected but the magnetic fluctuations are completely taken into account. Note, that the so-called "tree approximation" [3] does not yield the HLM effect and the self-consistent, or mean-field-like, approximation, mentioned above, is the simplest analytical method for an investigation of this phenomenon.

The present study is intended to provide enough theoretical results about the behaviour of measurable physical quantities directly related to the phase transition properties and in this way to ensure a theoretical basis for future experiments on the existence of the HLM effect. The need of an experimental observation of the HLM effect is very important because the effect remains a theoretical paradigm without a reliable experimental verification although its mechanism — the interaction of gauge fields in a quite universal Abelian–Higgs model — is of fundamental interest for different fields of physics as pure [5,9–13], and disordered [14–21] superconductors, quantum phase transitions [22,23], scalar electrodynamics [24], liquid crystals [25–29], and cosmology [30,31]. On the other hand, there are some theoretical studies, based on Monte Carlo simulations [32], the so-called dual model [33,34], and certain variants of the renormalization-group (RG) [35,36], in which no evidence of HLM effect was reported; for a discussion of this point, see the review article [37]. Therefore, in the modern theory of phase transitions the problem for the existence of HLM effect is controversial and cannot be easily solved without a hint from the experiment. The experimental research of the effect in liquid crystals cannot be considered reliable although the reported results are in favor of its existence.

Recently, it has been shown [9] that the HLM effect is stronger in quasi-2D superconducting films than in bulk superconductors and the preliminary evaluation of the relevant physical quantities like the order parameter jump and the latent heat at the equilibrium point of the fluctuation-induced first order transition in superconducting films gives for them several orders bigger

values than for those in bulk materials [10–13]. This result reopens the problem for an experimental search of HLM effect in type I superconductors, in particular, in thin films of type I superconductors with relatively small GL parameter κ. Here we shall investigate this problem in a comprehensive way.

We shall neglect the fluctuations of the superconducting order parameter because their effect on the thermodynamics of the superconductor is very weak; see, e. g., Refs. [2,3]. Within this approximation, the problems in the scope of the work can be considered without the use of RG, as well as of numerous and quite interesting RG results available in the literature; for a review, see, e. g., Ref. [37].

The approximation used by us does not allow for total solution of the long standing problem for the order of the phase transition in superconductors in a zero magnetic field. But we present a reliable investigation of the HLM effect outside the asymptotic vicinity of the phase transition point. Let us clarify this point in more details. We cannot address our investigation to certain classes of high-temperature superconductors where the (Ginzburg) critical region is quite large and the effect of the superconducting fluctuations should be taken into account. Besides, our studi is not intended to an investigation of the phase transition properties in the very narrow critical region ($|T - T_{c0}| \sim 10^{-12} - 10^{-16}$ K) in the usual low temperature superconductors with critical temperatures $T_{c0} < 20$ K. We concentrate our attention to phase transition properties which can be observed by the available experimental techniques, and for this reason we ignore phenomena which may occur in extremely small and, hence, experimentally unaccessible temperature intervals such as the Ginzburg critical region in usual superconductors. To ignore the critical region is equivalent to ignore the superconducting fluctuations by the supposition that their effect is negligibly small. Except for certain high-temperature superconductors, this approximation is comprehensive to all known superconducting systems and we shall use it in our consideration.

It seems at first sight that an obvious disadvantage of our investigation is that it cannot be included in the series of interesting recent works performed mainly by RG methods and intended to solve the problem for the effect of the superconducting fluctuations on the order of the superconducting phase transition in the asymptotic scaling region in the very vicinity of the critical point T_{c0}, i. e., to the check of the HLM effect in a regime of strong fluctuation interactions of the superconducting order parameter. However, the present work is not addressed to such aims. Rather, our efforts are concentrated on the investigation of the HLM effect in experimentally accessible temperature intervals ($\Delta T > 10^{-5}$ K) outside the negligibly small critical region, where the superconducting fluctuations in low-temperature superconductors can be ignored and the HLM effect due to the magnetic fluctuations can be experimentally tested. Therefore, we can reliably work within the mean-field-like approximation described in Ref. [1] and discussed in recent Refs. [9–13]. Besides, here we shall consider the same method in details together with the limits of its validity. Thus we shall demonstrate that our consideration of the HLM effect is valid outside the critical region. More detailed effects, as those predicted by RG and characterized by temperature scales of order 10^{-6} K and lower are beyond the scope of this article.

In Sec. II we present a derivation of the effective free energy of a D-dimensional superconductor. Subsections II.B and II.E are devoted to a detailed discussion of the validity of our approach. In Sec. III we give the first thorough investigation of the effective free energy for bulk superconductors. In Sec. IV the quasi-2D superconducting films and the validity of the Landau expansion are discussed. In Sec. V we summarize our main conclusions.

II. EFFECTIVE FREE ENERGY

A. Model considerations

The GL free energy [2] of a D-dimensional superconductor of volume $V_D = (L_1 ... L_D)$ is given in the form

$$F(\psi, \mathbf{A}) = \int d^D x \left[a|\psi|^2 + \frac{b}{2}|\psi|^4 + \frac{\hbar^2}{4m}\left| \left(\nabla - \frac{2ie}{\hbar c}\mathbf{A} \right) \psi \right|^2 + \frac{\mathbf{B}^2}{8\pi} \right] . \tag{1}$$

In Eq. (1) the first Landau parameter $a = \alpha_0(T - T_{c0})$ is expressed by the critical temperature $T_{c0} = T_c(H = 0)$ in a zero external magnetic field ($H = |\mathbf{H}|$), $b > 0$ is the second Landau parameter and $e \equiv |e|$ is the electron charge. The square B^2 of the magnetic induction $\mathbf{B} = (\mathbf{H} + 4\pi\mathbf{M})$, is given by the vector potential $\mathbf{A}(\mathbf{x}) = \{A_j(\mathbf{x}),\ j = 1, ..., D\}$ in the form

$$\mathbf{B}^2 = \frac{1}{2} \sum_{i,\,j\,=\,1}^{D} \left(\frac{\partial A_j}{\partial x_i} - \frac{\partial A_i}{\partial x_j} \right)^2 , \tag{2}$$

here the vector potential $\mathbf{A}(\mathbf{x})$ obeys the Coulomb gauge $\nabla \cdot \mathbf{A}(\mathbf{x}) = 0$. For a 3D superconductor the relation $\mathbf{B} = [\nabla \cdot \mathbf{A}(\mathbf{x})]$ can be used and when $\mathbf{B} = \mathbf{B}_0$ is uniform along the z-axis, the Landau gauge $\mathbf{A}_0(\mathbf{x}) = B_0(-y/2, -x/2, 0)$ can be applied. This representation can be generalized for $(D > 2)$-dimensional systems, where the magnetic induction B_0 is a second rank tensor:

$$B_{0ij} = B_0(\delta_{i1}\delta_{j2} - \delta_{j2}\delta_{i1}). \tag{3}$$

If we use the notation $\mathbf{x} = (x_1, x_2, \mathbf{r})$, where $\mathbf{r}$ is a $(D-2)$-dimensional vector perpendicular to the plane (x_1, x_2), in the 3D case we shall have $\mathbf{r} = (0, 0, z)$, and

$$B_j = \frac{1}{2}\epsilon_{jkl}B_{0kl} = B_0\delta_{j3} , \tag{4}$$

where ϵ_{jkl} is the antisymmetric Levi-Civita symbol. The Landau gauge and Eqs. (3)-(4) can be used for uniform $\mathbf{B} = \mathbf{B}_0$ when $\delta\mathbf{B}$ fluctuations are neglected; see, e. g. Ref. [6]. In the prevailing part of our study we shall apply the general Coulomb gauge of the field $\mathbf{A}(\mathbf{x})$ which does not exclude spatial dependent magnetic fluctuations $\delta\mathbf{B}(\mathbf{x})$.

In nonmagnetic superconductors where the mean value $\langle\mathbf{M}\rangle = (\mathbf{M} - \delta\mathbf{M})$ of the magnetization $\mathbf{M}$ is equal to zero in the normal state in zero external magnetic field, the magnetic induction in presence of external magnetic field takes the form:

$$\mathbf{B} = \mathbf{H}_0 + \delta\mathbf{H}(\mathbf{x}) + 4\pi\delta\mathbf{M}(\mathbf{x}) \ , \tag{5}$$

where $\mathbf{H}_0$ is the (uniform) regular part of the external magnetic field and $\delta\mathbf{H}$ is an irregular part of $\mathbf{H}$ created by uncontrollable effects. We neglect the irregular part $\delta\mathbf{H}$ and set $\mathbf{H}_0 = 0$, then $\mathbf{B}$ contains only a fluctuation part $\mathbf{B} \equiv \delta\mathbf{B}(\mathbf{x}) = 4\pi\delta\mathbf{M}(\mathbf{x})$ that describes the diamagnetic variations of $\mathbf{M}(\mathbf{x})$ around the zero value $\langle\mathbf{M}\rangle = 0$ due to fluctuations $\delta\psi(\mathbf{x})$ of the ordering field $\psi(\mathbf{x})$ above ($T > T_{c0}$) and below ($T < T_{c0}$) the normal-to-superconducting transition at T_{c0}. Note, that the nonfluctuation part $\mathbf{A}_0 = [\mathbf{A}(\mathbf{x}) - \delta\mathbf{A}(\mathbf{x})]$ corresponds to the regular part $\mathbf{B}_0 = (\mathbf{H}_0 + \langle\mathbf{M}\rangle) = 0$ of $\mathbf{B}$ in nonmagnetic superconductors ($\langle\mathbf{M}\rangle = 0$) in a zero external magnetic field ($\mathbf{H}_0 = 0$). Then we can set $\mathbf{A}_0(\mathbf{x}) = 0$ and, hence, $\delta\mathbf{A}(\mathbf{x}) = \mathbf{A}(\mathbf{x})$, so we have an entirely fluctuation vector potential $\mathbf{A}(\mathbf{x})$ which interacts with the order parameter $\psi(\mathbf{x})$. This interaction can be of type $\psi^2 A$ and $\psi^2 A^2$ and generates all effects discussed in the paper.

We accept periodic boundary conditions for the superconductor surface. This means to ignore the surface energy including the additional energy due to the penetration of the magnetic field in a surface layer of thickness equal to the London penetration depth $\lambda(T) = \lambda_0|t_0|^{-1/2}$, $t_0 = |T - T_{c0}|/T_{c0}$; $\lambda_0 = (mc^2 b/8\pi e^2 \alpha_0 T_{c0})^{1/2}$ is the zero-temperature value of λ. This approximation is adequate for superconductors of thickness $L_0 \gg \lambda(T) \gg a_0$, where a_0 is the lattice constant and $L_0 = \min\{L_i,\, i = 1, ..., D\}$. As we suppose the external magnetic field to be zero ($H_0 = 0$) or very small in real experiments, the requirement $L_0 \gg \lambda(T)$ cannot be satisfied and we take into account only the condition $L_0 \gg a_0$.

In microscopic models of periodic structures the periodic boundary conditions confine the wave vectors $\mathbf{k}_i = \{k_i = (2\pi n_i/L_i);\ i = 1, ..., D\}$ in the first Brillouin zone $[-(\pi/a_0) \le k_i < (\pi/a_0)]$ and the expansion of their values beyond this zone can be made either by neglecting the periodicity of the crystal structure or on the basis of the assumption that big wave numbers $k = |\mathbf{k}|$ have a negligible contribution to the calculated quantities. The last argument is widely accepted in the phase transitions theory where the long-wavelength ($ka_0 \ll 1$) limit can be used. In particular, this argument is valid in the continuum limit ($V_D/a_0^D \to \infty$). Therefore, for both crystal and nonperiodic structures we can use a cutoff $\Lambda \sim (\pi/a_0)$ and afterwards to extend this cutoff to infinity provided the main contributions in the summations over $\mathbf{k}$ come from the relatively small wavenumbers ($k \ll \Lambda$). Note, that here we make a quasimacroscopic description based on the GL functional (1) which means that the microscopic phenomena are excluded from our consideration.

The GL free energy functional takes into account phenomena with characteristic lengths ξ_0 and λ_0 or larger (ξ and λ) where $\lambda(T)$ is the London penetration length mentioned above and $\xi(T) = \xi_0|t|^{-1/2}$ is the coherence length [2]; here $\xi_0 = (\hbar^2/4m\alpha_0 T_{c0})^{1/2}$ is the zero-temperature coherence length. In low-temperature superconductors ξ_0 and λ_0 are much bigger than the lattice constant a_0. Having in mind this argument we shall assume that in our investigation $\Lambda \ll (\pi/a_0)$. Whether the upper cutoff Λ is chosen to be either $\Lambda \sim 1/\xi_0$ or $\Lambda \sim 1/\lambda_0$ is a problem that has to be solved by additional arguments (see Sec. III.C).

We shall use the Fourier expansion

$$A_j(\mathbf{x}) = \frac{1}{V_D^{\frac{1}{2}}} \sum_k A_j(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{x}} \tag{6}$$

and

$$\psi(\mathbf{x}) = \frac{1}{V_D^{\frac{1}{2}}} \sum_k \psi(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{x}} \ , \tag{7}$$

where the Fourier amplitudes $A_j(\mathbf{k})$ obey the relation $A_j^*(\mathbf{k}) = A_j(-\mathbf{k})$ and $\mathbf{k}\cdot\mathbf{A}(\mathbf{k}) = 0$. The Fourier amplitude $\psi(\mathbf{k})$ is not equal to $\psi^*(-\mathbf{k})$ because $\psi(\mathbf{x})$ is a complex function. For the same reason $\psi(0) \equiv \psi(\mathbf{k} = 0)$ is a complex number.

B. Approximations

The total ignoring of both superconducting and magnetic fluctuations in Eq. (1) leads to the familiar tree approximation where the GL equations [2] should be solved. Note, that the tree, or mean-field, approximation is the lowest order theory within the framework of the loop expansion, e. g., see [3,38]. The systematic treatment of the fluctuation effects in the asymptotic vicinity of the phase transition point can be given by RG.

The effect of the superconducting fluctuations $\delta\psi(\mathbf{x})$ on the phase transition properties is restricted in a negligibly small vicinity ($|t_0| \sim 10^{-12} \div 10^{-16}$) of the temperature T_{c0} and we shall assume that $\delta\psi(\mathbf{x}) = 0$, i. e., $\psi \approx \langle\psi(\mathbf{x})\rangle$; from now on we shall denote $\langle\psi(\mathbf{x})\rangle$ by ψ. So we apply a mean-field approximation with respect to the order parameter $\psi(\mathbf{x})$. Within this approximation we shall take into account the $\delta\mathbf{A}(\mathbf{x})$-fluctuations for $\mathbf{B}_0 = 0$, i. e., $\mathbf{A}(\mathbf{x}) = \delta\mathbf{A}(\mathbf{x})$. Furthermore, the $\mathbf{A}(\mathbf{x})$-fluctuations can be integrated out from the partition function, defined by:

$$Z(\psi) = \int \mathcal{D}A\, e^{-F(\psi,\mathbf{A})/k_B T} \ , \tag{8}$$

where the functional integral $\int \mathcal{D}A$ is defined by

$$\int_{-\infty}^{\infty} \prod_{j=1}^{D} \prod_{x \in V_D} dA_j(\mathbf{x}) \delta[\mathrm{div}\mathbf{A}(\mathbf{x})] \ . \tag{9}$$

The integration is over all possible configurations of the field $\mathbf{A}(\mathbf{x})$; the δ-function takes into account the Coulomb gauge.

The partition function $Z(\psi)$ corresponds to an effective free energy $\mathcal{F}$

$$\mathcal{F}_D = -k_{\mathrm{B}} T \ln Z(\psi) \ , \tag{10}$$

The magnetic fluctuations will be completely taken into account, if only we are able to solve exactly the integral (8). The exact solution can be done for a uniform order parameter ψ. The uniform value of ψ is different from the mean-field value of ψ because the uniform fluctuations of $\psi(\mathbf{x})$ always exist, so we should choose one of these two possibilities. The problem for this choice arises after the calculation the integral (8) at a next stage of consideration when the effective free energy $\mathcal{F}_{\mathcal{D}}$ is analyzed and the properties of the superconducting phase ($\psi > 0$) are investigated. The effective free energy is a particular case of the effective thermodynamic potential in the phase transition theory [3,38] and we must treat the uniform ψ in the way prescribed in the field theory of phase transitions. It will become obvious from the next discussion that we shall use a loop-like expansion which can be exactly summed up to give a logarithmic dependence on $|\psi|^2$.

Because of the spontaneous symmetry breaking of the continuous symmetry in the ground state, the ordered phase $\psi > 0$, i.e., the effective free energies discussed in this paper depend on the modulus $|\psi|$ of the complex number $\psi = |\psi| e^{i\theta}$ but not on the phase angle θ which remains arbitrary. That is why we shall consider the modulus $|\psi|$ as an "effective order parameter" because the angle θ does not play any role in the phenomena investigated in the paper. The quantity $|\psi|$ remains undetermined up to the stage when we define the equilibrium order parameter $|\psi_0|$ by the equation of state $[\partial \mathcal{F}_D(\psi)/\partial\psi] = 0$. This equation gives the equilibrium value ψ_0 of ψ and the difference $\delta\psi_0 = (\psi_0 - \psi)$ can be treated as the uniform (zero dimensional) fluctuation of the field $\psi(\mathbf{x})$. The $\mathbf{x}$-dependent fluctuations $\delta\psi(\mathbf{x})$ have been neglected because of the uniformity of ψ. The solution ψ_0 will be stable towards the uniform fluctuation $\delta\psi$ provided the same solution $\psi_0 = |\psi_0| e^{i\theta_0}$ corresponds to a stable (normal or superconducting) phase; the phase angle θ_0 remains unspecified. Therefore, we begin our investigation setting ψ uniform but at some stage of consideration we shall also ignore the uniform fluctuation $\delta\psi$ and deal only with the equilibrium value ψ_0 of ψ. The equilibrium value will be calculated after taking into account magnetic fluctuations, so it will be different from the usual result $|\psi_0| = (|a|/b)^{1/2}$ [2] when both magnetic and superconducting fluctuations are ignored. This simplest approximation for the equilibrium value of ψ is obtained from the GL free energy (1) provided $e = 0$ and the gradient term is neglected. Hereafter we shall keep the symbol $|\psi_0|$ for the equilibrium order parameter in the more general case when the magnetic fluctuations are not neglected and shall denote the same quantity for $e = 0$ by $\eta \equiv |\psi_0(e = 0)| = (|a|/b)^{1/2}$.

The above described approximation neglects the saddle point solutions of GL equations, where $\langle\psi(\mathbf{x})\rangle$ is $\mathbf{x}$-dependent. Therefore, the vortex state that is stable in type II superconductors cannot be achieved. This is consistent with the choice of a zero external magnetic field, where the vortex state cannot occur in any type superconductor. These arguments can be easily verified with the help of GL equations [2] for a zero external magnetic field; the only nonzero solution for ψ in this case is given by $\eta = (|a|/b)^{1/2}$ although the magnetic fluctuations $\mathbf{A}(\mathbf{x}) = \delta\mathbf{A}(\mathbf{x})$ are properly considered.

In conclusion we can argue that the described method will be convenient for both type I and type II superconductors in a zero external magnetic field, provided the ψ-fluctuations have a negligibly small effect on phase transition properties $T_{c0} = T_c(H_0 = 0)$, where T_c denotes the phase transition line for any $H_0 \geq 0$. For type II superconductors in $H_0 > 0$, two lines $T_{c1}(H_0)$ and $T_{c2}(H_0)$ should be defined, usually given by $H_{c1}(T)$ and $H_{c2}(T)$ [2].

C. Derivation of effective free energy

When the order parameter ψ is uniform the functional (1) is reduced to

$$F(\psi, \mathbf{A}) = F_0(\psi) + F_A(\psi) \ , \tag{11}$$

with

$$F_0(\psi) = V_D \left(a|\psi|^2 + \frac{b}{2}|\psi|^4 \right) \tag{12}$$

and

$$F_A(\psi) = \frac{1}{8\pi} \int d^D x \left\{ \rho(\psi) \mathbf{A}^2(\mathbf{x}) + \frac{1}{2} \sum_{i,j=1}^{D} \left(\frac{\partial A_j}{\partial x_i} - \frac{\partial A_i}{\partial x_j} \right)^2 \right\} \ . \tag{13}$$

Here $\rho = \rho_0 |\psi|^2$ and $\rho_0 = (8\pi e^2/mc^2)$. It is convenient to calculate the partition function $Z(\psi)$ and the effective free energy $\mathcal{F}_{\mathrm{D}}(\psi)$ in the $\mathbf{k}$-space, where Eqs. (9) and (13) take the form

$$\int_{-\infty}^{\infty} \prod_{j=1}^{D} \prod_{\mathbf{k}>0}^{k \leq \Lambda} d\mathrm{Re}A_j(\mathbf{k}) d\mathrm{Im}A_j(\mathbf{k}) \delta\left[\mathbf{k} \cdot \mathbf{A}(\mathbf{k})\right] \tag{14}$$

and

$$F_A(\psi) = F_A(0) + \Delta F_A(\psi) \ . \tag{15}$$

Here

$$F_A(0) = \frac{1}{8\pi} \sum_{j,k} k^2 \left|A_j(\mathbf{k})\right|^2 \ , \tag{16}$$

and

$$\Delta F_A(\psi) = \rho \sum_{j,k} \left|A_j(\mathbf{k})\right|^2 \ ; \tag{17}$$

note, that we have used the Coulomb gauge $\mathbf{k}\cdot\mathbf{A}(\mathbf{k}) = 0$.

Then the partition function (8) will be

$$\mathcal{Z}(\psi) = e^{-F_0(\psi)/k_{\mathrm{B}}T}\, \mathcal{Z}_A(\psi) \ , \tag{18}$$

where

$$\mathcal{Z}_A(\psi) = \int \mathcal{D}A\, e^{-F_A(\psi)/k_{\mathrm{B}}T} \tag{19}$$

with $F_A(\psi)$ given by (15) and the functional integration is defined by the rule (14). With the help of Eqs. (10)–(19) the effective free energy $\mathcal{F}_D(\psi)$ becomes

$$\mathcal{F}_D(\psi) = F_0(\psi) + \mathcal{F}_f(\psi) \ , \tag{20}$$

where $F_0(\psi)$ is given by Eq. (12) and

$$\mathcal{F}_f(\psi) = -k_{\mathrm{B}}T \ln\left[\frac{\mathcal{Z}(\psi)}{\mathcal{Z}(0)}\right] \tag{21}$$

is the ψ-dependent fluctuation part of $\mathcal{F}(\psi)$. In Eq. (20) the ψ-independent fluctuation energy $\{-k_{\mathrm{B}}T \ln[\mathcal{Z}_A(0)]\}$ has been omitted. This energy should be ascribed to the normal state of the superconductor which, by convention, is set equal to zero.

Defining the statistical averages

$$\langle(\ldots)\rangle = \frac{\int \mathcal{D}\mathcal{A}\, e^{-F_A(0)/k_{\mathrm{B}}T}(\ldots)}{\mathcal{Z}_A(0)}, \tag{22}$$

we can write Eq. (21) in the form

$$\mathcal{F}_f(\psi) = -k_{\mathrm{B}}T \ln\langle e^{-\Delta F_A(\psi)/k_BT}\rangle. \tag{23}$$

Eq. (23) is a good starting point for the perturbation calculation of $\mathcal{F}_f(\psi)$. We expand the exponent in Eq. (23) and also take into account the effect of the logarithm on the infinite series [3] and obtain in result

$$\mathcal{F}_f(\psi) = \sum_{l=1}^{\infty} \frac{(-1)^l}{l!(k_{\mathrm{B}}T)^{l-1}} \langle \Delta F_A^l(\psi)\rangle_c \ , \tag{24}$$

where $\langle\ldots\rangle_c$ denotes connected averages [3]. Now we have to calculate averages of the type

$$\langle A_\alpha(\mathbf{k}_1), A_\beta(\mathbf{k}_2)\ldots A_\gamma(\mathbf{k}_n)\rangle_c \ . \tag{25}$$

Here we shall use the Wick theorem and the correlation function of form

$$G_{ij}^{(A)}(\mathbf{k},\mathbf{k}') = \langle A_i(\mathbf{k}) A_j(-\mathbf{k}')\rangle = \delta_{\mathbf{k},\mathbf{k}'} G_{ij}^A(k) \ , \tag{26}$$

where

$$G_{ij}^A(k) = \langle A_i(k) A_j(-k)\rangle = \frac{4\pi k_{\mathrm{B}}T}{k^2}\left(\delta_{ij} - \hat{k_i}\hat{k_j}\right) \tag{27}$$

and $\hat{k_i} = (k_i/k)$.

The calculation of lowest order terms ($l = 1, 2, 3$) in Eq. (24) with the help of (25)–(27) is straightforward. The infinite series (24) can be exactly summed up and the result is the following logarithmic function

$$\mathcal{F}_f(\psi) = \frac{(D-1)}{2}\, k_{\mathrm{B}}T \sum_k \ln\left[1 + \frac{\rho(\psi)}{k^2}\right] \ . \tag{28}$$

The same result for $\mathcal{F}_f(\psi)$ can be obtained by a direct calculation of the Gaussian functional integral (9). This is done using the integral representation of δ-function in (9) or (14) but it introduces an additional functional integration that should be carried out after the integration over $A_j(\mathbf{x})$.

Eqs. (10), (20) and (28) give the effective free energy density

$$f_D(\psi) = \mathcal{F}_D(\psi)/V_D \tag{29}$$

in the form

$$f_D(\psi) = f_0(\psi) + \Delta f_D(\psi) \ , \tag{30}$$

where

$$f_0(\psi) = a|\psi|^2 + \frac{b}{2}|\psi|^4 \tag{31}$$

and

$$\Delta f_D(\psi) = \frac{(D-1)k_{\mathrm{B}}T}{2V_D} \sum_k \ln\left(1 + \frac{\rho}{k^2}\right) \ . \tag{32}$$

Eqs. (20) and (29)–(32) are the basis of our further considerations. We should mention that the fluctuation contribution $\Delta f_D(\psi)$ to $f(\psi)$ transforms to a convergent integral in the continuum limit

$$\frac{1}{V_D}\sum_k \to \int \frac{d^D k}{(2\pi)^D} = K_D \int_0^\Lambda dk\, k^{D-1} \ , \tag{33}$$

where $K_D = 2^{1-D}\pi^{-D/2}/\Gamma(D/2)$ for all spatial dimensionalities $D \geq 2$. But the terms in the expansion of the logarithm in (32) are power-type divergent with the exception of several low-order terms in certain dimensionalities D. Therefore, we shall work with a finite sum of an infinite series of infinite terms. In our further calculations we shall keep the cutoff Λ finite for all relevant terms in $\Delta f_D(\psi)$. This is the condition to obtain correct results.

D. Particular dimensions

For purely 2D superconductor consisting of a single atomic layer, we can use Eqs. (29)–(32) setting $D = 2$ and calculate $\Delta f_2(\psi)$ with the help of the rule (33):

$$\Delta f_2(\psi) = \left(\frac{k_{\rm B}T}{8\pi}\right)\left[(\Lambda^2 + \rho_0|\psi|^2)\ln\left(1+\frac{\rho_0|\psi|^2}{\Lambda^2}\right) - \rho_0|\psi|^2\ln\left(\frac{\rho_0|\psi|^2}{\Lambda^2}\right)\right] . \quad (34)$$

The first term of this free energy can be expanded in powers of $|\psi|^2$:

$$\Delta f_2(\psi) = \left(\frac{k_{\rm B}T}{8\pi}\right)\{\rho_0|\psi|^2 + \rho_0|\psi|^2\ln\left(\frac{\Lambda^2}{\rho_0|\psi|^2}\right) + \frac{\rho_0^2|\psi|^4}{2\Lambda^2}\} . \quad (35)$$

Thus we obtain the result from Ref. [39]. This case is of special interest because of the logarithmic term in the Landau expansion for $f(\psi)$ but it has no practical application for the lack of ordering in purely 2D superconductors.

For quasi-2D superconductors we assume that $(2\pi/\Lambda) > L_0 \gg a_0$, where L_0 is the thickness of the superconducting film and a more precise choice of the upper cutoff $\Lambda \ll (1/a_0)$ for the wave numbers k_i is a matter of an additional investigation [9] (see Sec. II.A and II.E). In order to justify this definition of a quasi-2D system we consider the more general case of a 3D system of volume $V = (L_1 L_2 L_0)$, where we can take the continuum limit along the large dimensions (L_1 and L_2) of the film because of the assumption $L_\alpha \gg (2\pi/\Lambda)$, $(\alpha = 1, 2)$. The summation over the wave number $k_0 = (2\pi n_0/L_0)$ cannot be substituted with an integration because $L_0 \ll L_\alpha$ and the dimension L_0 does not obey the conditions, valid for L_α [40–42]. Therefore, for such 3D system we must sum over k_0 and integrate over two other components (k_1 and k_2) of the wave vector $\mathbf{k}$. This gives an opportunity for a systematic description of the 2D–3D crossover in superconductors [13,41–44] which fully justifies the application of more simple treatment for $a_0 \ll L_0 < (2\pi/\Lambda)$.

We consider the conditions $(2\pi/\Lambda) > L_0 \gg a_0$ as a definition of a quasi-2D films of thickness L_0. The condition $(2\pi/\Lambda) > L_0$ means that the sum in Eq. (32) contains only terms with $(k_0 = 0)$. The summation over $\mathbf{k} = (k_1, k_2, 0)$ gives a correct description of quasi-2D films of thickness L_0 and this can be shown as a limiting case of the more general 2D–3D crossover described in Refs. [13,41–44]. Therefore, for a quasi-2D film we have the expression;

$$\Delta f(\psi) = \frac{2}{L_0}\Delta f_2(\psi) , \quad (36)$$

where $\Delta f_2(\psi)$ is given by Eq. (34).

For the bulk (3D) superconductor we obtain:

$$\Delta f_3(\psi) = \frac{k_B T}{2\pi}\left[\frac{\Lambda^3}{3}\ln\left(1+\frac{\rho_0|\psi|^2}{\Lambda^2}\right) + \frac{2}{3}\rho_0|\psi|^2\Lambda - \frac{2}{3}\rho_0^{3/2}|\psi|^3\arctan\left(\frac{\Lambda}{\sqrt{\rho_0|\psi|^2}}\right)\right] . \quad (37)$$

For the Landau expansion in powers of $|\psi|$ this form of $f_3(\psi)$ confirms the respective results in Refs. [1,4] and moreover correctly gives a term of type $\rho_0^2|\psi|^4$ which was supposed small and neglected in these preceding papers. This problem will be discussed in Sec. III.

For 4D-systems $\Delta f_D(\psi)$ becomes

$$\Delta f_4(\psi) = \frac{3k_{\rm B}T}{64\pi^2}\left[\Lambda^2\rho_0|\psi|^2 + \Lambda^4\ln\left(1+\frac{\rho_0|\psi|^2}{\Lambda^2}\right) - \rho_0^2|\psi|^4\ln\left(1+\frac{\Lambda^2}{\rho_0|\psi|^2}\right)\right] . \quad (38)$$

The above expression for $\Delta f_4(\psi)$ can be also expanded in powers of $|\psi|$ to show that it contains a term of the type $|\psi|^4\ln(\sqrt{\rho_0}|\psi|/\Lambda)$ which produces a first order phase transition; this case is considered in the scalar electrodynamics [24]. In our further investigation we shall focus our attention on 3D and quasi-2D superconductors.

The free energy density $\Delta f_{\rm D}(\psi)$ can be expanded in powers of $|\psi|$ but the Landau expansion can be done only in an incomplete way for even spatial dimensions. Thus $f_2(\psi)$, $f_4(\psi)$, and $f(\psi)$ being the free energy density corresponding to the quasi-2D films, contain logarithmic terms which should be kept in their original

form in the further treatment of the function $\Delta f_D(\psi)$ in the Landau expansion. We shall do our analysis in two ways: with and without Landau expansion of $\Delta f_D(\psi)$. These variants of the theory will be called "exact" theory (ET) and "Landau" theory (LT), respectively. We shall show that these two ways of investigation give the same results in all cases except for quasi-2D films with relatively small thicknesses ($L_0 \ll \xi_0$). It seems important to establish the differences between two variants of the theory because the HLM effect is very small and any incorrectness in the theoretical analysis may be a cause for an incorrect result. By same arguments we shall investigate the effect of the factor T in $\Delta f_D(\psi)$ on the thermodynamics of quasi-2D films. This factor can be represented as $T = T_{c0}(1 + t_0)$ and one may expect that the usual approximation $T \approx T_{c0}$, which is well justified in the Landau theory of phase transitions [2,3], may be applied. We shall show for both 3D and quasi-2D superconductors, that this way of approximation can be made by neglecting terms in the thermodynamic quantities smaller than the leading ones. On the other hand, practical calculations lead to the conclusion that this approximation cannot be made without a preliminary examination because for some quasi-2D films it produces a substantial error of about 10%. LT, in which the factor T is substituted by T_{c0}, will be called a "simplified Landau expansion" (SLT).

E. Validity

The general result (29)–(32) for the effective free energy $f(\psi)$ has the same domain of validity [2] as the GL free energy functional in a zero external magnetic field. When we neglect a sub-nano interval of temperatures near the phase transition point we can use Eq. (1) provided $|t_0| = |T - T_{c0}|/T_{c0} < 1$, or in the particular case of type I superconductors, $|t_0| < \kappa^2$ [2]. Note, that the latter inequality does not appear in the general GL approach. It comes as a condition for the consistency of this approach with the microscopic BCS theory for type I superconductors [2].

Taking the continuum limit we have to assume that all dimensions of the body, including the thickness L_0, are much larger than the characteristic lengths ξ and λ. The exception of this rule is when we consider thin films. Especially for thin films of type I superconductors, where $((2\pi/\Lambda) > L_0 \gg a_0)$, we should have in mind that $\xi(T) > \lambda(T)$, so the inequalities $\xi > \lambda > \xi_0 > \lambda_0$ hold true in the domain of validity of the GL theory $|t_0| < \kappa^2 < 1$. In Ref. [9] a comprehensive choice of the cutoff Λ has been made ($\Lambda = \xi_0$) and we shall discuss this point in Sec. 3 and 4. Note, that the respective conditions for quasi-2D films of type II superconductors are much weaker and are reduced to the usual requirements: $\kappa > 1/\sqrt{2}$, $|t_0| < 1$ and $(2\pi/\Lambda) > L_0 \gg a_0$.

If we do a Landau expansion of $f_D(\psi)$, in powers of $|\psi|^2$ the condition $\rho \ll \Lambda^2$ should be satisfied. In order to evaluate this condition we substitute $|\psi|^2$ in $\rho = \rho_0|\psi|^2$ with $\eta^2 = |a|/b$ which corresponds to $\epsilon = 0$ (Sec. II.B). As $\lambda^2(T) = 1/\rho$, the condition for the validity of the Landau expansion becomes $[\Lambda\lambda(T)]^2 \gg 1$, i.e., $(\Lambda\lambda_0)^2 \gg |t_0|$. Choosing the general form of $\Lambda_\tau = (\pi\tau/\xi_0)$ where τ describes the deviation of Λ_τ from $\Lambda_1 \equiv \Lambda = (\pi/\xi_0)$, we obtain $(\pi\tau\kappa)^2 \gg |t_0|$; $\kappa = (\lambda_0/\xi_0)$ is the GL parameter.

Thus we can conclude that in type II superconductors, where $\kappa = (\lambda_0/\xi_0) > 1/\sqrt{2}$, the condition $(\rho/\Lambda^2) \ll 1$ is satisfied very well for values of the cutoff in the interval between $\Lambda = (\pi/\xi_0)$ and $\Lambda = (\pi/\lambda_0)$, i.e., for $1 < \tau < (1/\kappa)$. For type I superconductors, where $\kappa < 1/\sqrt{2}$ the cutoff values $\Lambda \sim (1/\xi_0)$ leads to the BCS condition ($|t_0| < \kappa^2$) for the validity of the GL approach. Substantially larger cutoffs ($\Lambda \gg \pi/\xi_0$), for example, $\Lambda \sim (1/\lambda_0)$ for type I superconductors with $\kappa \ll 1$ lead to a contradiction of this BCS condition with the requirement $\rho \ll \Lambda^2$. This inconsistency will be discussed again in Sec. II.C.

In our calculations we often use another parameter $\mu_\tau = (1/\pi\tau\kappa)^2$ and, in particular, $\mu \equiv \mu_1 = (1/\pi\kappa)^2$ and in terms of μ the condition for the validity of expansion of $f_D(\psi)$ becomes $\mu|t_0| \ll 1$, or, more generally, $\mu_\tau|t_0| \ll 1$. Choosing $\tau = 1/\pi$ we obtain the BCS criterion for the validity of the GL free energy of type I superconductors [2]. The choice $\tau = (\xi_0/\pi\lambda_0)$ corresponds to the cutoff $\Lambda_\tau = 1/\lambda_0$. As we shall see in Sec. III and IV the thermodynamics near the phase transition point has no substantial dependence on the value of the cutoff Λ_τ but it should be chosen in a way that is consistent with the mean-field-like approximation.

Alternatively, the inequality $(\rho/\Lambda^2) \ll 1$ may be investigated with the help of the reduced order parameter φ defined by $\varphi = |\psi|/\eta_0$, where $\eta_0 \equiv \eta(T = 0) = (\alpha_0 T_{c0}/b)^{1/2}$ is the so-called zero-temperature value of the order parameter within the GL free energy $f_0(\psi)$, given by Eq. (31); see also Sec. II.B. The reduced order parameter φ will be equal to $|t_0|$ for $t_0 < 0$, if only the magnetic fluctuations are ignored, i.e., when $|\psi| = \eta$. Using the notation φ, we obtain the condition $(\rho/\Lambda^2) \ll 1$ in the form $\mu_\tau\varphi^2 \ll 1$. This condition seems to be more precise because it takes into account the effect of magnetic fluctuations on the order parameter ψ.

III. BULK SUPERCONDUCTORS

A. Free energy

The effective free energy $f_3(\psi)$ of bulk (3D-) superconductors is given by Eqs. (29)–(31) and (37). The analytical treatment of this free energy can be done by Landau expansion in small $(\sqrt{\rho_0}|\psi|/\Lambda)$. Up to order $|\psi|^6$ we obtain

$$f_3(\psi) \approx a_3|\psi|^2 + \frac{b_3}{2}|\psi|^4 - q_3|\psi|^3 + \frac{c_3}{2}|\psi|^6 \,, \qquad (39)$$

where

$$a_3 = a + \frac{k_B T \Lambda \rho_0}{2\pi^2} \,, \qquad (40)$$

$$b_3 = b + \frac{k_B T \rho_0^2}{2\pi^2 \Lambda} , \tag{41}$$

$$q_3 = \frac{k_B T \rho_0^{3/2}}{6\pi} , \tag{42}$$

and

$$c_3 = -\frac{k_B T \rho_0^3}{6\pi^2 \Lambda^3} . \tag{43}$$

The cutoff Λ in Eqs. (40)–(43) is not specified and can be written in the form $\Lambda_\tau = (\pi\tau/\xi_0)$ as suggested in Sec. 2.5.

We shall just outline the analysis of the above free energy. It can be shown by both analytical and numerical calculations [10] that $|\psi|^6$-term has no substantial effect on the thermodynamics, described by the free energy (39). That is why we ignore this term and do the analysis in the standard way [3]. The possible phases $|\psi_0|$ are found as a solution of the equation of state:

$$[\partial f(\psi)/\partial|\psi|\,]_{\psi_0} = 0 . \tag{44}$$

There always exists a normal phase $|\psi_0| = 0$ which gives a minimum of $f_3(\psi)$ for $a_3 > 0$. The possible superconducting phases are given by

$$|\psi_0|_\pm = \frac{3q_3}{4b_3}\left(1 \pm \sqrt{1 - \frac{16 a_3 b_3}{9 q_3^2}}\right) \geq 0. \tag{45}$$

Having in mind the existence and stability conditions of $|\psi_0|_\pm$-phases [3], we obtain that the $|\psi_0|_+$-phase exists for $(16 a_3 b_3) \leq 9 q_3^2$ and this region of existence always corresponds to a minimum of $f_3(\psi)$. The $|\psi_0|_-$-phase exists for $0 < a_3 < (9q_3^2/16 b_3)$ and this region of existence always corresponds to a maximum of $f_3(\psi)$, i.e., this phase is absolutely unstable. For $a_3 = 0$, $|\psi_0|_- = 0$ and hence, coincides with the normal phase. For $9q_3^2 = (16 a_3 b_3)$ we have $|\psi_0|_+ = |\psi_0|_- = (3q_3/4b_3)$ and $f_3(|\psi_0|_+ = f_3|\psi_0|_-) = (27 q_3^4/512 b_3^3)$. Furthermore, $f_3(|\psi_0|_-) > 0$ for all allowed values of $|\psi_0|_- > 0$, whereas

$$f_3(|\psi_0|_+) < 0 \quad \text{for} \quad a_3 < (q_3^2/2b_3) ,$$

and

$$f_3(|\psi_0|_+) > 0 \quad \text{for} \quad (q_3^2/2b_3) < a_3 < \frac{9q_3^2}{16 b_3} .$$

The equilibrium temperature $T_{\rm eq}$ of the first order phase transition is defined by the equation $f(|\psi_0|_+) = 0$ which gives the following result:

$$2 b_3(T_{\rm eq}) a_3(T_{\rm eq}) = q_3^2(T_{\rm eq}) . \tag{46}$$

These results are confirmed by numerical calculations of the effective free energy (39) [10]; there also the influence of the $|\psi|^6$-term is evaluated.

B. Entropy and specific heat capacity

The equilibrium entropy jump is $\Delta S = V\Delta s$ and $\Delta s = -(df_3(|\psi|)/dT)$ can be calculated with the help of Eq. (39) and the equation of state (44):

$$\Delta s = -|\psi_0|^2 \Phi(|\psi_0|) , \tag{47}$$

where $\Phi(|\psi_0|)$ is the following function:

$$\Phi(y) = \left(\alpha_0 + \frac{k_B \Lambda \rho_0}{2\pi^2}\right) - \frac{\rho_0^{3/2} k_B}{6\pi} y + \left(\frac{k_B \rho_0^2}{4\pi^2 \Lambda}\right) y^2 . \tag{48}$$

The specific heat capacity per unit volume $\Delta C = T(\partial \Delta s/\partial T)$ is obtained from (47)

$$\Delta C = -\left(\frac{T}{T_{c0}}\right) \frac{\partial |\psi_0|^2}{\partial t_0} \Phi(|\psi_0|) . \tag{49}$$

The quantities $\Delta s(T)$ and $\Delta C(T)$ can be evaluated at the equilibrium phase transition point $T_{\rm eq}$ which is found from Eq. (46):

$$\frac{T_{\rm eq}}{T_{c0}} \approx 1 - \frac{k_B \rho_0 \Lambda}{2\pi^2 \alpha_0} + \frac{\left(\rho_0^{3/2} k_B/6\pi\right)^2}{b + (\rho_0^2 k_B/2\pi^2\Lambda)\, T_{c0}} \left(\frac{T_{c0}}{\alpha_0}\right) , \tag{50}$$

provided $|\Delta T_c| = |T_{c0} - T_{\rm eq}| \ll T_{c0}$. Further we shall see that the condition $|\Delta T_c| \ll T_{c0}$ is valid in real substances. The second term in r.h.s. of Eq. (50) is a typical negative fluctuation contribution whereas the positive third term in r.h.s. of the same equality is typical for first-order transitions [3].

To obtain the jumps Δs and ΔC at $T_{\rm eq}$ we have to put the solution $|\psi_0|_+$ found from Eq. (45) in Eqs. (47)–(49). The result will be:

$$\Delta s = -\frac{q_{3c}^2}{b_{3c}^2}\left\{\alpha_0 + \frac{k_B \rho_0 \Lambda}{2\pi^2} - \left(\frac{k_B \rho_0^{3/2}}{6\pi}\right)^2 \frac{T_{\rm eq}}{b_{3c}^2}\right\} , \tag{51}$$

and

$$\Delta C = \frac{4\alpha_0}{b_{3c}}\left(\alpha_0 T_{c0} - \frac{q_{3c}^2 b}{b_{3c}^2}\right) , \tag{52}$$

where b_{3c} and q_{3c} are the parameters b_3 and q_3 at $T = T_{\rm eq}$. As $|\Delta T_c| = |T_{c0} - T_{\rm eq}| \ll T_{c0}$ we can set $T_{\rm eq} \approx T_{c0}$ in r.h.s. of Eqs. (51) and (52) and obtain $q_{3c} \equiv q_3(T = T_{\rm eq}) \approx q_3(T_{c0})$ and $b_{3c} \approx b_3(T_{c0})$.

The latent heat $Q = -VT_{\rm eq}\Delta s$ of the first order phase transition at $T_{\rm eq}$ can be calculated from Eq. (51). If we neglect the charge ($e = 0$) which means to set

$\rho_0 = q_3 = 0$ and $b_{c3} = b$ in Eqs. (51)–(52) we shall get the result from Ref. [1] for the ratio

$$(\Delta T)_{\rm eq} = \frac{Q}{T_{\rm eq}\Delta C} \ . \qquad (53)$$

Here we should mention that Eq. (52) gives the jump ΔC at the equilibrium phase transition point of the first order phase transition, described by $|\psi|^3$ term [3], while ΔC calculated in Ref. [1] is equal to the specific heat jump at the standard second order transition $\Delta C = (\alpha_0^2 T_{c0}/b)$ and is four times smaller. Therefore, we obtain $(\Delta T)_{\rm eq}$ four times smaller than the respective value in Ref. [1].

C. Numerical values for Al

In order to do the numerical estimates we represent the Landau parameters α_0 and b with the help of the zero-temperature coherence length ξ_0 and the zero-temperature critical magnetic field H_{c0}. The connection between them is given by formulae of the standard GL theory of superconductivity [2]: $\xi_0^2 = (\hbar^2/4m\alpha_0 T_{c0})$ and $H_{c0}^2 = (4\pi\alpha_0^2 T_{c0}^2/b)$. The expression for the zero-temperature penetration depth $\lambda_0 = (\hbar c/2\sqrt{2}eH_{c0}\xi_0)$ is obtained from the above relation and $\lambda_0 = (b/\alpha_0 T_{c0}\rho_0)^{1/2}$. We shall use the following experimental values of T_{c0}, H_{c0} and ξ_0 for Al: $T_{c0} = 1.19$ K, $H_{c0} = 99$ Oe, $\xi_0 = 1.6\,\mu$m, $\kappa = 0.01$ [1,45]. The experimental values for T_{c0}, H_{c0} and ξ_0 vary about 10–15% depending on the method of measurement and the geometry of the samples (bulk material or films) but such deviations do not affect the results of our numerical investigations.

The evaluation of the parameters a_3 and b_3 for Al gives:

$$a_3 = (\alpha_0 T_{c0})\left[t_0 + 0.972\times 10^{-4}(1+t_0)\tau\right], \qquad (54)$$

and

$$\frac{b_3}{b} = 1 + \frac{0.117}{\tau} \ . \qquad (55)$$

Setting $\tau = 1$ corresponds to the cutoff $\Lambda_1 = (\pi/\xi_0)$ (Sec 2.5). For $\tau = (1/\kappa)_{\rm Al} = 10^2$ which corresponds to the much higher cutoff $\Lambda = (\pi/\lambda_0)$ we have $b_3 \approx b$, i. e., the ρ_0^2 -term in b_3, given by Eq. (41), can be neglected. However, as we see from Eq. (55), for $\tau = 1$ the same ρ_0^2-correction in the parameter b_3 is of order $0.1b$ and cannot be automatically ignored in all calculations, in contrast to the supposition in Refs. [1,4]. However, the more important fluctuation contribution in 3D superconductors comes from the τ-term in Eq. (54) for the parameter a_3. This term is of order 10^{-4} for $\tau \sim 1$ and this is consistent with the condition $|t_0| < \kappa^2 \sim 10^{-4}$ but for $\tau \sim 10^2$, i. e., for $\Lambda \sim (\pi/\lambda_0) \sim 10^6\,\mu$m, the same $\tau-$ term is of order 10^2 which exceeds the temperature interval $(T_{c0} \pm 10^{-4})$ for the validity of BCS condition of Al (Sec. II.E).

These results demonstrate that for our theory to be consistent, we must choose the cutoff $\Lambda_\tau = (\pi\tau/\xi_0)$, where τ is not a large number ($\tau \to 1 \div 10$). To be more concrete we set $\Lambda = \Lambda_1 = (\pi/\xi_0)$ as suggested in Ref. [9].

The temperature shift $t_{\rm eq} = t_0(T_{\rm eq})$ for bulk Al can be estimated with the help of Eq. (50). We obtain that this shift is negative and very small: $t_{\rm eq} \sim -10^{-4}$. Note, that the second term in the r.h.s. of Eq. (50) is of order 10^{-4} provided $\Lambda \sim (1/\xi_0)$ whereas the third term in the r.h.s. of the same equality is of order 10^{-5}. Once again the change of the cutoff Λ to values much higher than (π/ξ_0) will take the system outside the temperature interval where the BCS condition for Al is valid. Let us note, that in Ref. [10] the parameter t corresponds to our present notation t_0. But the numerical calculation of the free energy function $f_3(\psi)$ in Ref. [10] was made for the SLT variant of the theory and the shifted parameter $(t_0 + 0.972\times 10^{-4})$ was incorrectly identified with t and this lead to the wrong conclusion for its positiveness at the equilibrium phase transition point $T_{\rm eq}$. As a matter of fact, the shifted parameter $(t_0 + 0.972\times 10^{-4})$ is positive at $T_{\rm eq}$ but $t_{\rm eq} \equiv t_0(T_{\rm eq})$ is negative.

Having in mind these remarks, when we evaluate Δs and ΔC for bulk Al we can use simplified versions of (51) and (52) which means to consider only the first terms in the r.h.s and to take $q_{3c} \approx q_3$ and $b_{3c} \approx b$ at T_{c0}. In this way we obtain

$$Q = -T_{c0}\Delta s = 0.8\times 10^{-2}\left[\frac{\rm erg}{\rm K\cdot cm^3}\right] \ , \qquad (56)$$

and

$$\Delta C = 2.62\times 10^3\left[\frac{\rm erg}{\rm cm^3}\right] \ . \qquad (57)$$

The results are consistent with an evaluation of ΔC for Al as a jump $(\Delta\tilde{C} = \alpha_0^2 T_{c0}/b)$ at the second order superconducting transition point [1] that, as we mentioned above, is four times smaller than the jump ΔC given by Eq. (57).

A complete numerical evaluation of the function $f_3(\psi)$ and the jump of the order parameter at $T_{\rm eq}$ for bulk Al was presented for the first time in Ref. [10]. The results there confirm that the order parameter jump and Q for bulk type I superconductors are very small and can hardly be observed in experiments.

We shall finish the presentation of bulk Al with a discussion of the ratio (53). It can be also written in the form

$$(\Delta T)_{\rm eq} = \frac{32\pi}{9}\left(\frac{T_{c0}^2}{b\alpha_0}\right)\left(\frac{e^2}{mc^2}\right)^3 , \qquad (58)$$

and it differs by a factor 1/4 from the respective result in Ref. [1]. This difference is due to the fact that we take ΔC as the jump at the first order transition temperature $T_{\rm eq}$ while in the above cited paper [1] the authors define ΔC as a hypothetic jump $(\Delta\tilde{C})$ at the standard second order phase transition point. From Eq. (56) we obtain

$$(\Delta T)_{\rm eq} = 6.7\times 10^{-12}(T_c^3 H_{c0}^2 \xi_0^6), \qquad (59)$$

and multiplying the number coefficient in the above expression by 4 we can obtain Eq. (10) from Ref. [1].

IV. QUASI-2D FILMS

Thin quasi-2D films ($a_0 \ll L_0 < 2\pi/\Lambda$) can be investigated with the help of the respective free energy density $f(\psi)$ given by Eqs. (30) and (31), $\Delta f_D(\psi)$ is taken from Eqs. (34) and (36). The free energy of quasi-2D superconducting films was derived and analyzed for the first time in Ref. [9] using the Landau expansion of $\Delta f_2(\psi)$ in powers of $|\psi|^2$; see Eq. (35). As is shown for the first time by Lovesey [39] in the simple 2D case the fluctuation contribution $\Delta f_{\rm D}(\psi)$, of form given by Eq. (35), leads to a fluctuation-induced first order phase transition. In contrast to 3D superconductors where the first order of the phase transition is generated by $|\psi|^3$-term in $\Delta f_3(\psi)$, in 2D superconductors the first order of the phase transition is a result of the presence of $|\psi|^2\ln|\psi|$ in Eq. (35). But the Meissner phase cannot occur in 2D (single atomic layer) superconductors because of the strong fluctuations and hence this case is of no interest. In quasi-2D films, where the Meissner phase does occur for properly chosen thickness of the film ($L_0 \ll 2\pi/\Lambda$) [9], the change of the order of normal-to-superconducting phase transition is better pronounced than in bulk superconductors. This is well illustrated in the above cited paper [9] by numerical data for Al films with thickness $L_0 = 0.1$ μm. Following Refs. [9,11] and the arguments presented in Sec. III.C we shall choose the cutoff $\Lambda = \pi/\xi_0$.

The expansion of the respective free energy in powers of $|\psi|$ leads to somewhat clumsy analysis and for this reason we shall use the approach in Ref. [11,12] where the quasi-2D films have been investigated with the help of the general form of $\Delta f(\psi)$ described by Eq. (34). In both variants of the theory (ET and LT; see Sec. II.D) the thickness L_0 of the quasi-2D film has an effect on the thermodynamic behaviour, that is similar to the influence of material parameters α_0 and b. This is very well seen in the Landau expansion (35) of the free energy $f(\psi)$ given by (36), where the parameters a and b acquire a fluctuation contribution that depends on L_0. The influence of L_0 on the thermodynamic properties can be considered as a characteristic feature of quasi-2D systems [42,43], a feature, absent in purely 2D films [39]. It is unambiguously demonstrated by several theoretical studies of the 2D–3D crossover in systems with slab geometry [13,42–44] that the L_0-dependence as given in Eq. (36) correctly describes quasi-2D films.

Following Refs. [11,12] and having in mind the above discussion we can present the free energy density $f(\psi) = (F(\psi)/L_1L_2)$ in the form

$$f(\varphi) = \frac{H_{c0}^2}{8\pi}\left[2t_0\varphi^2 + \varphi^4 + C(1+t_0)\Gamma(\mu\varphi^2)\right], \qquad (60)$$

where

$$\Gamma(y) = (1+y)\ln(1+y) - y\ln y, \qquad (61)$$

$$C = \left(\frac{2\pi^2 k_{\rm B} T_{c0}}{L_0\xi_0^2 H_{c0}^2}\right). \qquad (62)$$

In Eqs. (60)–(62) we have set $\Lambda = (\pi/\xi_0)$ and introduced the notation $\varphi = |\psi|/\eta_0$; the quantity η_0 is defined in Sec. II.E. Some of the properties of free energy (60) were analyzed in Ref. [11] for Al films and in Ref. [12] for films of Tungsten (W), Indium (In), and Aluminium (Al). Here we shall summarize and justify the preceding results and, moreover, we shall present new results about the properties of the Landau expansion of effective free energy. Note, that the function $\Gamma(y)$ cannot be fully expanded in powers of y because of the term of type $(y\ln y)$ in Eq. (61).

The extensive investigations [11,12] of films of W, Al and In with thicknesses from 0.05 μm to 2 μm confirm the intuitive notion that the HLM effect is stronger for smaller values of L_0. The numerical analysis shows that type I superconductors with relatively small GL parameter κ and relatively high critical field H_{c0} may be the best candidates for the experimental observation of the effect. The best material from the above enumerated substances seems to be Al; tungsten has an extremely small GL parameter but also a small critical field that makes it inconvenient for experiments. The relatively high H_{c0} of In results in relatively large latent heat, $Q \sim 4.0$ (erg/cm^3) but for films with $L_0 \sim 0.05$ μm the order parameter jump $|\psi|_{\rm eq} = \varphi_{\rm eq}\eta_0$ at $T_{\rm eq}$ is twice smaller than that for the respective Al films [12]: $|\psi|_{\rm eq} = 0.05 \times 10^{-11}$ for In and $|\psi|_{\rm eq} = 0.1 \times 10^{-11}$ for Al. We have to stress the role of critical magnetic field H_{c0}, a fact established for the first time in [12] and the present paper. With the help of data from Refs. [9–12] we compared the thermodynamic quantities near the first order phase transition point in bulk Al and Al films of $L_0 \sim 0.1$ μm. They are given in Table 1, where $t_{\rm eq} = t_0(T_{\rm eq})$, $\varphi_{\rm eq} = \varphi(T_{\rm eq})$ is the equilibrium jump of the reduced order parameter and $|\psi|_{\rm eq} = \varphi_{\rm eq}\eta_0$ is the order parameter jump at $T_{\rm eq}$.

quantity	$t_{\rm eq}$	$\varphi_{\rm eq}$	$\lvert\psi\rvert_{\rm eq}$	Q (erg/cm^3)
bulk Al	-0.492×10^{-4}	0.0032	0.8×10^{9}	0.8×10^{-2}
$L_0 = 0.1$ μm	-0.00147	0.032	0.8×10^{10}	0.8

Table 1. Numerical data for bulk Al and Al film of thickness $L_0 = 0.1$ μm.

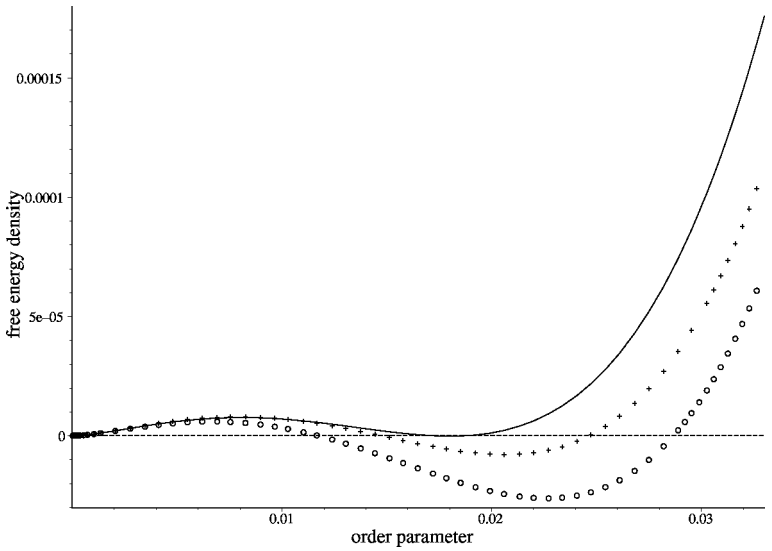

Fig. 1. The function $f(\varphi)$ for Al films of thickness $L_0 = 0.4$ μm: the solid line corresponds to ET, the line of crosses (+) represents LT, and the line of circles (∘) stands for SLT. All curves are calculated for $T_{\rm eq}$ corresponding to ET (see the text).

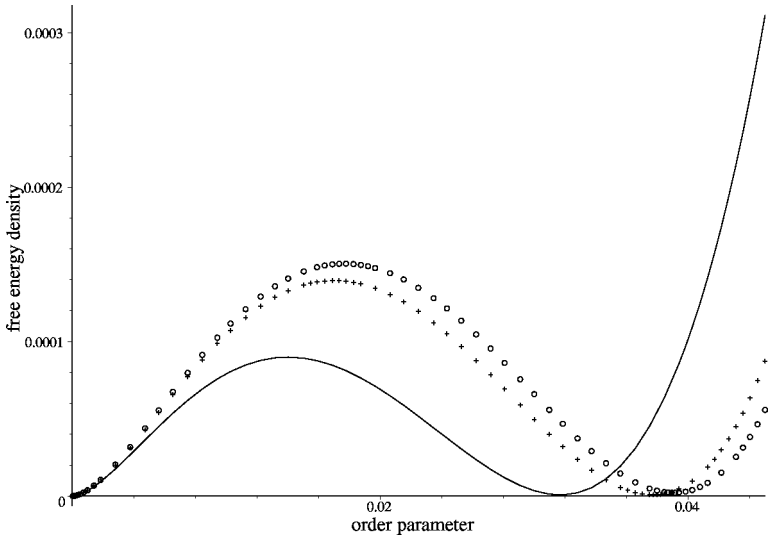

Fig. 2. The function $f(\varphi)$ for Al films of thickness $L_0 = 0.1$ μm: the solid line (–) corresponds to ET, the line of crosses (+) represents LT, and the line of circles (∘) stands for SLT. All curves are calculated for $T_{\rm eq}$ corresponding to the respective variant of the theory (see the text).

The shift $t_{\rm eq}$ of the equilibrium transition temperature due to magnetic fluctuations is very small in both bulk Al and thin Al films so the difference $(T_{\rm eq} - T_{c0})$ can be neglected in all calculations of thermodynamic quantities near $T_{\rm eq}$. The equilibrium jump $\varphi_{\rm eq}$, or, equivalently, $|\psi|_{\rm eq}$, is one order of magnitude higher in the film with $L_0 = 0.1$ μm than in bulk Al but the latent heat Q is 10^2 times bigger for films. These values are almost one order of magnitude higher for $L_0 \sim 0.05$ μm than for $L_0 = 0.1$ μm. The numerical data in Table 1 are obtained by SLT for the bulk Al samples and by ET for the Al film of thickness $L_0 = 0.1$ μm; for the abbreviations SLT, LT and ET, see Sec. II.D. The difference in the numerical results obtained from ET, LT and SLT will be discussed in the remainder of the paper.

The investigation of bulk superconductors yields the same results irrespective of whether we analyze the free energy $f_3(\psi)$ by ET, LT or SLT. The situation in quasi-2D superconductors is however different; the three different variants of treatment of the free energy give different results, in particular, for relatively small thicknesses $(L_0 \ll \xi_0)$. This feature of free energy, Eq. (60), is il-

lustrated in Fig. 1, where three curves for three different variants of $f(\psi)$ are shown for Al films of thickness $L_0 = 0.4$ μm. The solid line corresponds to ET, the line of crosses represents the LT result, and the line of circles stands for SLT. All three curves are calculated for $t_{\rm eq} \equiv t_0(T_{\rm eq}) = -0.00057$, which is the ET equilibrium phase transition temperature $T_{\rm eq} = 0.9994\ T_{c0}$. Note, that $\varphi_{\rm eq}$ is the nonzero global minimum of $f(\varphi)$ and the function $f(\varphi)$ depicted in Fig. 1 has only one minimum for $\varphi > 0$ because all curves are calculated in the thermodynamic regime corresponding to the stable Meissner phase.

The main conclusion that can be made from Fig. 1 is that the two variants of the Landau expansion give approximately the same quantitative results and therefore, the factor $(1+t_0)$ in (60) can always be substituted by unity, though the present investigation is intended to quite small physical effects. This conclusion is consistent with the argument [11] that allows to use the same approximation $(1+t_0) \approx 1$ for the calculation of Q and ΔC in both variants of the theory: ET and LT. Besides, the Fig. 1 shows that both variants of LT give slightly higher equilibrium phase transition temperatures $T_{\rm eq}$ and substantially higher equilibrium jumps $\varphi_{\rm eq}$ than ET. Thus, using LT for film thicknesses $a_0 \ll L_0 < 0.1$ μm one may obtain up to 10 times higher value of $\varphi_{\rm eq}$ and up to 10^2 times bigger latent heat Q than the respective values in Table 1. The problem is whether these higher values predicted by LT are reliable.

Fig. 2 shows the free energy drawn in the three variants (ET, LT and SLT) of $f(\varphi)$ for Al films of thickness $L_0 = 0.1$ μm. In Fig. 2, the curves $f(\varphi)$ are drawn at their respective equilibrium phase transition points. The variation in $t_{\rm eq}$ (-0.00148 for the solid line, -0.00115 for "+" -line and -0.00046 for $\circ$-line) are of order of the typical values of $t_{\rm eq}$ itself so the differences in $t_{\rm eq}$ due to the way of calculation cannot be neglected. Although for both variants of the Landau expansion (LT and SLT), the quantity $\varphi_{\rm eq}$ is again practically the same, the difference in $t_{\rm eq}$ is more pronounced for smaller thickness of Al film and moreover, both variants of Landau expansion are not so good approximation to the result of the exact calculation (ET) as for $L_0 = 0.4$ μm. The conclusions, we have already drawn from the results shown in Fig. 1, are completely confirmed by the form of the curves from Fig. 2 and, moreover, we see that the deviation of the results of LT from those of ET becomes bigger with the decrease of film thickness L_0.

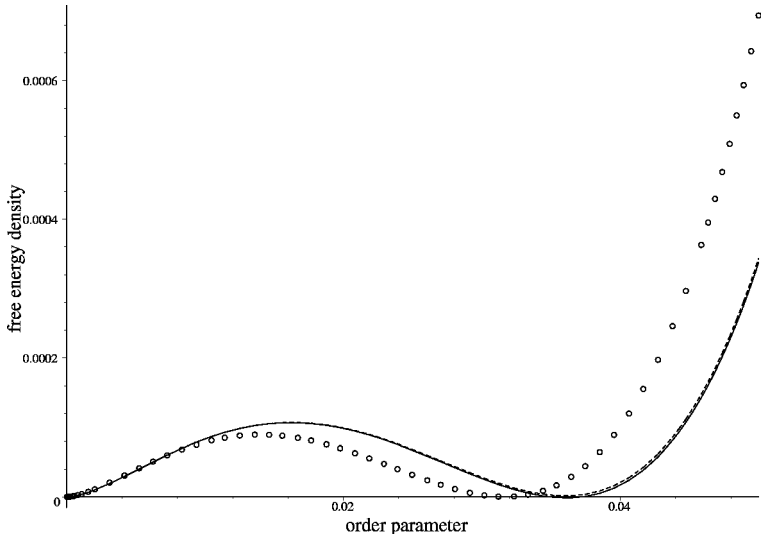

Fig. 3. The function $f(\varphi)$ calculated from the ET for Al films of thickness $L_0 = 0.1$ μm and different cutoffs Λ: the solid line (–) corresponds to $\Lambda = \pi/\lambda_0$, the dashed line represents the case $\Lambda = 1/\lambda_0$, and the line of circles ($\circ$) stands for $\Lambda = \pi/\xi_0$. All curves are calculated at the respective $T_{\rm eq}$ (see the text).

On the basis of the above observations we may conclude that in all cases when ET and the respective Landau expansions give different results, the Landau expansion yields a better established first order transition, with a higher jump $\varphi_{\rm eq}$, and hence, bigger values of Q and ΔC. In order to establish where LT is a good approximation we have made systematic numerical calculations for Al films of different thicknesses $L_0 = 0.05 \div 3$ μm. When L_0 is lowered beginning with 3 μm the quantitative differences between the two variants of theory, with and without Landau expansion, respectively, become substantial about $L_0 \approx 0.4$ μm. Bearing in mind the condition for the validity of the Landau expansion (see, Sec. II.E) and the requirement for the equilibrium jump of the order parameter, $\mu\varphi_{\rm eq}^2 < 1$ we may suppose that the predictions done with the help of the Landau expansions do not satisfy this inequality for Al films with thicknesses $L_0 \leq 0.1$ μm. For this relatively small L_0-size, ET is absolutely reliable. The numerical data show that films with $L_0 > 0.1$ μm are described well quantita-

tively by the Landau expansion. The differences between the curves in Figs. 1 and 2 can be neglected in numerical calculations intended to give theoretical predictions for experiments.

This general result is supported by the following simple argument. In the Landau expansion of free energy (60) for quasi-2D films the parameter a acquires a cutoff (Λ-) independent contribution of the form

$$\Delta a = \frac{k_{\rm B} T \rho_0}{4\pi L_0} \,. \tag{63}$$

When we compare Δa with the bare parameter value $|a| = (\alpha_0 T_{c0} |t_0|)$ for Al, it is easily obtained that $|t_0|$ will not exceed 10^{-4} for thicknesses L_0 which are of order 1 μm or larger. Therefore, for $L_0 \sim 0.1$ μm the Landau expansion gives results which are quantitatively different from those obtained by ET.

We have studied the dependence of the free energy density $f(\varphi)$ of Al films with $L_0 = 0.1$ μm on the cutoff Λ. Fig. 3 shows the free energy density $f(\varphi)$ for three values of the cutoff: $\Lambda = \pi/\lambda_0$, $\Lambda = 1/\lambda_0$, and $\Lambda = \pi/\xi_0$. As the cutoff increases from (π/ξ_0) to $\pi/\lambda_0 \sim 10^2 (\pi/\xi_0)$, the equilibrium jump $\varphi_{\rm eq}$ increases, too. We have already mentioned in Sec. III.C that the increase of the cutoff Λ for type I superconductors up to the value (π/λ_0) is inconsistent in the present theory. The numerical result for Al films shown in Fig. 3 is, therefore, a demonstration of the validity of our arguments about the choice of the cutoff Λ presented in Sec. III.C. If we take the cutoff $\Lambda \gg (\pi/\xi_0)$ we shall go beyond the scope of validity of our theory.

There is a similarity between the breakdown of the present theory for cutoffs $\Lambda \gg (\pi/\xi_0)$ and the breakdown of the condition $(\rho/\Lambda^2) \ll 1$ for the validity of LT at small thicknesses L_0. In both cases, when there is an inconsistency of the theory, we obtain enhanced values of the characteristic jumps of thermodynamic quantities at the equilibrium point of the first-order phase transition.

V. CONCLUSION

We did a detailed analysis of the HLM effect in bulk (3D-) superconductors and quasi-2D superconducting films within the self-consistent approximation introduced in Refs. [1,4,24]. We have studied for a first time the validity of this approximation and calculated thermodynamic quantities of direct experimental interest like the equilibrium jumps of the order parameter, entropy and specific heat at the point of the fluctuation-induced first-order phase transition to superconducting state in a zero external magnetic field. Our investigation is supported by numerical calculations for bulk Al and Al films.

We have presented for a first time a comprehensive analysis of the effective free energy of the superconductor in a zero external magnetic field and on the basis of this analysis we compared the results from the investigation of the effective free energy without a partial Landau expansion with those from the Landau expansion of the effective free energy of quasi-2D films. For quasi-2D films of type I superconductors, the Landau expansion leads to reliable results, provided the film thickness is above some value depending on the characteristic lengths ξ_0 and λ_0, i. e., on the material parameters. For Al films the results from the Landau expansion become unreliable below film thicknesses $L_0 \sim 0.1 \mu$m. For quasi-2D films of type II superconductors the Landau expansion of the effective free energy can be used for any thickness above the level of destruction of superconductivity ($L_0 \sim 10^{-3}$ μm).

Our investigation provides a reliable theoretical basis for a future experimental search of the HLM effect in thin films of type I superconductors, where the effect is much stronger than in bulk materials. In accord with preceding works [9–13] we have justified earlier results which indicate that the HLM effect will be better pronounced in films of materials with relatively high values of the critical magnetic field H_{c0} and relatively small thicknesses L_0. We cannot be certain which superconducting material provides the best experimental conditions for transport or caloric measurements of the jumps of the thermodynamic quantities at the point of the fluctuation-induced first order transition, but from the data for Al, W and In available from recent studies [11,12] and the present paper, the most suitable substance seems to be Al. But investigations of other superconductors may put forward materials which are even better candidates for the experimental test of the HLM effect.

Looking for the most convenient material for the experimental search of the HLM effect we should have in mind a number of other experimental requirements which are not related to the results from the present theoretical investigation. Here we shall briefly discuss the problem of the external magnetic field and the possible change of the superconductivity from type I to type II with the decrease of the film thickness [46]. This change for film thicknesses L_0, which are convenient for the experimental study of the HLM effect, is not a great problem because we have shown in our investigation that the effect could be observed also in films of type II superconductors, provided the external magnetic field H_0 is very low so the effect of the vortex phase and the magnetic energy jump $(H_0^2/8\pi)$ at the phase transition point is negligible. The magnetic energy jump $(H_0^2/8\pi)$ may obscure the HLM effect on the latent heat also in type I superconductors and, therefore the experimental problem for the elimination of the residual laboratory external magnetic field H_0 is common for both type I and type II superconducting films. If we take as a basis the latent heat of order 1 [erg/cm^3] in Al films with $L_0 \sim 0.1$ μm, as reported in Sec. 4, the magnetic field which ensures the ratio $(H_0^2/8\pi Q) \ll 1$ will be obviously about 1 Oe. In thinner films of convenient materials this experimental condition may become $H_0 \sim 10$ Oe but no more.

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НОВІ РЕЗУЛЬТАТИ ВПЛИВУ МАГНЕТНИХ ФЛЮКТУАЦІЙ НА ФАЗОВИЙ ПЕРЕХІД У НАДПРОВІДНИЙ СТАН ПРИ ВІДСУТНОСТІ МАГНЕТНОГО ПОЛЯ

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Подано систематичне дослідження впливу магнетних флюктуацій на фазовий перехід у надпровідний стан при відсутності зовнішнього магнетного поля за допомогою самоузгодженого наближення і квазімакроскопічної моделі Ґінзбурга–Ландау. Отримано нові результати для тонких надпровідних плівок. Розглянуто цікаві з експериментального погляду термодинамічні величини, такі, як стрибок параметра порядку, латентна теплота, теплоємність; їх чисельно розраховано для об'ємних Al та тонких Al плівок. Проаналізовано можливість експериментальної перевірки теоретичних передбачень.