Fluctuation induced first order phase transition in thin films of type I superconductors

R. Folk\textsuperscript{a}, D. V. Shopova\textsuperscript{b,*}, D. I. Uzunov\textsuperscript{a−c}

\textsuperscript{a}Institut für Theoretische Physik, Johannes Kepler Universität Linz, A-4040 Linz, Austria
\textsuperscript{b}CPCM Laboratory, G. Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences, BG-1784 Sofia, Bulgaria
\textsuperscript{c}Max-Planck-Institut für Komplexer Systeme, 01187 Dresden, Germany

*Corresponding author: sho@issp.bas.bg

Key words: superconductivity, magnetic fluctuations, latent heat, specific heat, order parameter profile.

PACS: 74.20.-z, 64-30+t, 74.20.De

Abstract

The effect of fluctuation induced weakly first order phase transition known for three dimensional (3D) type I superconductors appears in a modified and strongly enhanced variant in thin (quasi-2D) superconducting films. The unusual thermodynamic properties of this new type of first order phase transitions and the possibility for an experimental verification of the effect are established and discussed.

One of the outstanding problems of the phase transition theory of superconductivity is the so-called fluctuation induced weakly first order phase transition (WFOT) in three dimensional (3D) type I superconductors. It was shown \cite{1} that the interaction between the fluctuations $\delta H$ of the magnetic field $H$ and the order parameter $\psi(x)$ may produce a fluctuation induced change of the order of the phase transition in 3D type I superconductors: from a second order phase transition when the magnetic fluctuations $\delta H$ are neglected to a weak first order transition when the same magnetic fluctuations are taken into account and the mean magnetic field $H_0 = (H - \delta H)$ is equal to zero. Note, that we consider nonmagnetic superconductors, where the magnetic induction $B$ is equal to the magnetic field $H$. Besides, we discuss the phase transition point $T_{c0}$ corresponding to a
zero critical magnetic field, \( H_c(T_c) = 0 \), and our consideration will be not extended to the entire line \([0 \leq H_c(T) \leq H_c(0)]\) of the superconducting phase transition.

The same type of fluctuation induced first order phase transition was predicted also in other fields of physics, for example, in the theory of the early universe \([2, 3]\) and on the basis of the de Gennes model of smectic A – nematic phase transitions in liquid crystals \([4, 5]\). It seems that under certain circumstances, i.e. suitable values of the characteristic lengths, such fluctuation induced first order transitions may occur in any system described by an Abelian-Higgs type of model \([4, 6, 5]\), where a scalar field like the superconducting order parameter \( \psi(x) \) interacts with a massless vector gauge field. In a superconductor the massless gauge field is the vector potential \( A(x) \) of the magnetic field given for 3D systems by \( H(x) = \nabla \times A \) and the Coulomb gauge \( \text{div} A = 0 \). A general description of the phase transition mechanism in Abelian-Higgs systems was given within the scalar field electrodynamics \([3]\). The theoretical studies of this problem in superconductors have been performed with the help of the Ginzburg-Landau (GL) theory of superconductivity and the so-called dual model \([1, 11]\), including the application of the renormalization group (for a review, see \([8, 9]\)). The WFOT problem seems to be of general interest to a wide range of systems and, in particular, to recent aspects of superconductivity \([12, 13]\). According to the theoretical predictions the effect in 3D superconductors is very weak \([1, 7]\). For this reason an experimental search of the WFOT was attempted in smectic A liquid crystals by interface velocity measurements as well as by high-resolution heat capacity and X-ray experiments \([14, 15]\). The results were interpreted \([14, 15]\) in favour of a WFOT due to the interaction between the smectic scalar order parameter and the director vector of the nematic order, i.e. as a verification of the theoretically established fluctuation mechanism.

We shall restrict our consideration to type I superconductors where the description of the WFOT can be given within the "approximation of spatially uniform order parameter" \( (\psi(x) \approx \psi \ [1, 7]) \). That is why we shall present a form of the GL free energy which can be used for the investigation of the WFOT in D-dimensional type I superconductors for a uniform order parameter \( \psi \). The 3D and 4D dimensionalities were analyzed in preceding works \([1, 6]\) and we shall focus on quasi-2D films. We shall demonstrate that the effect of WFOT is much stronger in films than in 3D samples and, therefore, could be experimentally observed (if it really exists). Besides, in quasi-2D superconductors the WFOT is described by a logarithmic term instead of a third order term in \( \psi \ [1, 8]\), and this presents a special theoretical interest.

In standard notations \([10]\) the GL free energy in case of a spatially uniform order parameter \( \psi \) takes the form \( F_{GL} = (F_0 + F_A) \) with

\[
F_0 = V \left( a|\psi|^2 + \frac{b}{2} |\psi|^4 \right), \tag{1}
\]

and

\[
F_A = \frac{1}{16\pi} \sum_{ij} \int d^Dx \left[ 2\rho(\psi)\delta_{ij}A^2_j + (\partial_i A_j - \partial_j A_i)^2 \right], \tag{2}
\]
where $\partial_i = \partial/\partial_t$, $(i,j) = 1, \ldots, D$; $V = (L_1 \ldots L_D)$ is the volume of the D-dimensional superconductor, $H = |H|$, $a = \alpha_0(T - T_{c0})$ with $\alpha_0(T_{c0}) > 0$, $b(T_{c0}) > 0$, $\rho = \rho_0 |\psi|^2$ with $\rho_0 = (8\pi e^2/m_e^2)$. The quantities $2m$ and $2e$ are the effective mass and charge of the Cooper pairs respectively, and in superconductors of interest to the present investigation $2m \approx 2m_e$, where $m_e$ is the electron rest mass. The concrete values of the initial (bare) critical temperature $T_{c0}$ and the Landau parameters $\alpha_0$ and $b$ given by the microscopic BCS theory \cite{16}, are not essential for our consideration within the general GL phenomenological approach. But we should keep in mind that the GL theory describes only quasimacroscopic phenomena with characteristic lengths greater than the zero-temperature coherence length $\xi_0 = (\hbar^2/4m\alpha_0 T_{c0})^{1/2}$; for a microscopic (BCS) justification of this argument, see, e.g., \cite{10}.

In the space of the wave vectors $k$, the upper cutoff $A$ for $k \equiv k$ is $A \sim (1/\xi_0)$ and can be extended to larger values only in case of special circumstances (calculation of cutoff independent integrals). Besides, the free energy (1) has additional restrictions which are also relevant to our consideration. Firstly, the approximation $\psi = \text{const}$ will be valid for well established type I superconductors, like Al, where the GL parameter $\kappa$ is much less than unity; $\kappa = \lambda(T)/\xi(T)$. The coherence length $\xi(T)$ is given by $\xi(T) = \xi_0/|t|^{1/2}$, where $t = (T - T_{c0})/T_{c0}$, and the London penetration depth is $\lambda(T) = \lambda_0/|t|^{1/2}$, where $\lambda_0 = (mc^2 b/8\pi e^2 \alpha_0 T_{c0})^{1/2}$ stands for the zero-temperature value of $\lambda$. Secondly, we have to take into account the additional restriction on the GL theory for type I superconductors, namely, $|T - T_{c0}| \ll \kappa^2 T_{c0}$ \cite{10}, which comes from the general Landau condition $|T - T_{c0}| \ll T_{c0}$ and the requirement for a locality of the functional dependence between the supercurrent and the vector potential. The latter requirement is fulfilled for slow variations of the magnetic field and the vector potential at distances of order of $\xi_0$, i.e. for $\lambda(T) \gg \xi_0$. This means that the GL theory for well established type I superconductors with, for example, a GL parameter $\kappa \sim 10^{-1} \div 10^{-2}$, is valid only for $|t| < 10^{-2} \div 10^{-4}$ K. The Ginzburg critical region for these superconductors is very small ( $\sim 10^{-12} \div 10^{-16}$ K \cite{8,10}) and can be safely ignored. All phenomena that can be reliably predicted within this theory are confined to a relatively narrow vicinity of the phase transition point (0 $\sim 10^{-12} < |t| < 10^{-2} \div 10^{-4}$).

Thus we shall use the free energy (1) only for the description of phenomena in the temperature domain defined by the inequalities $\xi(T) \gg \lambda(T) \gg \xi_0$ which are fulfilled in the same vicinity of the phase transition point. Our heuristic arguments presented above have a reliable justification within the microscopic approach \cite{16}.

Following preceding papers \cite{1,3,17} we shall integrate out the vector potential in order to obtain an effective free energy as a function of the uniform field $\psi$. This integration is performed exactly within a simple one-loop expansion, as depicted in Fig.1, where the $\rho$-dependent term in eq. (2) is considered as a perturbation. The summation of the
infinite logarithmic series shown in Fig. 1 yields

\[ F^{(A)}_{\text{eff}} = \frac{1}{2}(D - 1)k_B T \sum_k \ln \left[ 1 + \rho(\psi)/k^2 \right] , \]

which substitutes \( F_A \) in \( F_{GL} \). The total effective free energy density is given by \( f_{\text{eff}} = (F_0 + F^{(A)}_{\text{eff}})/V \). The function \( f_{\text{eff}}(\psi) \) was investigated in details for \( D \geq 3 \) \[1\] \[2\] when the effects of the term \( F^{(A)}_{\text{eff}} \) are small; a brief notice \[17\] about the 2D case is also known.

Let us focus the attention on the quasi-2D spatial dimensionality (3D thin films), where the film thickness \( L_0 \) satisfies the condition \( \lambda(T) \gg L_0 \gg \xi_0 \). This condition is suitable for both theoretical and experimental investigations. Then our relatively thin superconducting slab must obey the following conditions:

\[ \xi(T) \gg \lambda(T) \gg L_0 \gg \xi_0 . \]

It is easy to see that the \( k \)--summation in eq. (3) can be substituted with a 2D integration over the wave vector components parallel to the film surface and the transverse wave vector component is equal to zero. An upper cutoff \( \Lambda = (1/\xi_0) \) is assumed for \( k \equiv |k| \).

In contrast to the pure 2D case \[17\] and studies in other spatial dimensionalities \[1\] \[4\] \[7\] the result for the effective free energy of the quasi-2D superconductor is given by

\[ f_{\text{eff}} = r_0 |\psi|^2 + \frac{u}{2} |\psi|^4 - Q |\psi|^2 \ln(\rho_0 |\psi|^2/\Lambda^2) , \]

where \( r_0 = (a + k_B T \rho_0/4\pi L_0) \), \( u = (b + k_B T \rho_0^2/4\pi \Lambda^2 L_0) \), and \( Q = (k_B T \rho_0/4\pi L_0) \). The evaluation of the contribution of the vector potential fluctuations, i.e. of the \( \rho_0 \)--dependent terms in eq. (5) shows that these terms cannot be neglected for the entire temperature interval of validity of our consideration. Besides, the evaluation of the new terms in the free energy is useful as a demonstration of the important role of the cutoff \( \Lambda \) and the necessity of the consistent choice \( \Lambda \approx (1/\xi_0) \). Another choice of \( \Lambda \) immediately leads to wrong predictions.

The free energy \( f(\varphi) = [f_{\text{eff}}(\varphi)/u] \) with \( \varphi \equiv |\psi| \geq 0 \) is depicted in Fig. 2 for several values of \( r = [r_0 + Q \ln(\Lambda^2/\rho_0)/u] \), and \( q = (Q/u) \). Figure 2 shows a well established first order phase transition. We shall briefly summarize the analytical treatment of the free energy (5) in terms of the simple notations \( f, r, \) and \( q \). The analysis is similar to that for first order phase transitions described by a third order \((\varphi^3-)\) term; for details, see \[8\]. Let us denote the solutions of the equation of state \( (\partial f/\partial \varphi) = 0 \) by \( \varphi_0 (\equiv |\psi_0|) \) - the possible phases. The normal phase solution \( (\varphi_0 = 0) \) exists and gives a minimum of \( f \) for all \( T \geq 0 \). Moreover, the necessary stability condition \( f''(\varphi_0) = (\partial^2 f/\partial^2 \varphi)_{\varphi_0} > 0 \) exhibits a singular behaviour \( f''(\varphi_0 \rightarrow 0) \rightarrow (+\infty) \), i.e. a logarithmic divergence. This peculiar property does not create troubles about the physical meaning of the effective free energy. Rather this result shows that in thin films the normal state can occur at any \( T \geq 0 \) as a stable phase above the equilibrium phase transition temperature \( T_c \neq T_{c0} \), or, if certain experimental conditions are satisfied, as a metastable phase for \( 0 \leq T \leq T_c \).
The superconducting (Meissner) phase \( \varphi_0 > 0 \) is described by the equation

\[
(r - q) + \varphi_0^2 - q \ln \varphi_0^2 = 0 .
\] (6)

The solution of this equation, i.e. the equilibrium order parameter \( \varphi_0 > 0 \) is shown in Fig. 3 for values of the parameters \( r \) and \( q \) which allow the existence of a first order transition. The check of the necessary \( (f''(\varphi_0) > 0) \) and the sufficient \( (f(\varphi_0) < 0) \) stability conditions for \( \varphi_0 > 0 \) show that the former is fulfilled for \( \varphi_0^2 > q \), whereas the latter condition is fulfilled for \( \varphi_0^2 > 2q \). Therefore, the Meissner phase can be overheated above the equilibrium transition temperature \( T_c \) of the phase transition defined by the equation \( \varphi_0^2(T_c) = 2q(T_c) \). The equilibrium order parameter jump is equal to \( (2q_c)^{1/2} \), where \( q_c \equiv q(T_c) \). This overheating may continue up to the temperature \( T_{c1} \) defined by \( \varphi_0^2(T_{c1}) = q(T_{c1}) \), where the order parameter becomes equal to \( q_c^{1/2} \) – the lowest nonzero value of the order parameter, for which it gives a minimum of \( f \). These features of the order parameter are shown in Fig. 3, where the ”metastability extension” of the order parameter profile is also drawn for a suitable choice of the parameter \( q \). Note, that one can easily show in an analytical way that stable solutions \( 0 < \varphi_0 < 1 \) of the eq. (6) always exist, at least, for \( r < 0 \).

The latent heat \( L(T_c) = T_c \Delta S(T_c) \) and the specific heat jump \( \Delta C(T_c) = T_c(\partial S/\partial T)_{T_c} \) at the equilibrium phase transition point \( T_c \) can be calculated from the free energy (5). We shall be interested in the temperature size of this first order transition, namely, in the ratio \( (\Delta T)_c = |L(T_c)/C(T_c)| \). Taking into account only the temperature dependence of the effective free energy \( f_{\text{eff}} \) on the Landau parameter \( a \), the temperature size \( (\Delta T)_c \) can be correctly evaluated. Our result is

\[
(\Delta T)_c = \frac{Q}{\alpha_0} ,
\] (7)

and after the substitution \( \alpha_0 = (h^2/4mT_c\xi_0^2)^{1/2} \) and \( Q = (k_BT_0/4\pi\lambda^2 L_0) \) we have

\[
(\Delta T)_c = 8k_BT_c^2 \left( \frac{\xi_0^2}{L_0} \right) \left( \frac{e}{hc} \right)^2 .
\] (8)

When we put in the above expression the numerical values of the fundamental constants and the tabulated data for Al \( (T_c = 1.19 \text{ K}, \) and \( \xi(0) = 1.6 \times 10^{-4} \text{ cm} ) \), we obtain that \( (\Delta T)_c \sim 10^{-6}/L_0 \). Note, that our approach is valid, if the thickness \( L_0 \) is greater than \( \xi_0 \) and less than \( \lambda(T) \). A choice of \( L_0 \) consistent with all theoretical requirements is \( L_0 \sim 10 \times \xi_0 \), or, for Al, \( L_0 \sim 10^{-3} \text{ cm} \), which yields \( (\Delta T)_c \sim 10^{-3} \text{ K} \). As the usual GL theory for Al is restricted within a temperature interval of size \( 10^{-4} \text{ K} \), our result indicates that the size of the WFOT covers totally the domain of validity of our unusual theory, namely we are faced with a normal size FOT rather than with a WFOT. Therefore, it should be emphasized that if the gauge mechanism of a fluctuation induction of the phase transitions order change really exists, this should be seen in suitable experiments with pure samples of good type I superconducting films of thickness about 10\( \mu \text{m} \). The
same result for the temperature size of the FOT can be obtained by the estimation of the parameter $\alpha_0$ given by the BCS theory [16].

In conclusion, we have three notes:

(1) Our approach to quasi-2D superconductors allows to use the 3D values of the original GL parameters $\alpha_0$ and $b$ and keeps the consideration far from dangerous effects of destruction of the order by 2D fluctuations. In this respect as well as in the values of the effective parameters $r_0$, $u$, and $Q$, our effective free energy (5) is quite different from the pure 2D free energy known from a preceding paper of Lovesey [17]. That is why we are able to make the prediction about the enhancement of the WFOT in quasi-2D superconductors (or, which is the same, ”almost 3D” superconductors). We should stress that for pure 2D case the Landau parameters $\alpha_0$ and $b$ are different from those corresponding to 3D superconductors and, hence, the evaluation of the same FOT effect in two dimensions needs a consistent treatment of these parameters. Moreover, the investigation of very thin (almost 2D) films requires an evaluation of the temperature region where the order parameter fluctuations will destroy the phase transition.

For these reasons our results cannot be extended in a straightforward way to very thin films by the respective decrease of the film thickness $L_0$. Within the phenomenological approach to dimensional crossover phenomena [18], the 3D Landau parameters are related to the 2D ones by $X_{(2D)} = \lim_{L_0 \to 0} (XL_0)$, where $X = (\alpha_0, b)$. This consideration should be compared with the BCS predictions for the 2D values of the free energy parameters.

(2) We have followed a method of integration of the magnetic fluctuations proposed in preceding papers [1, 5], in which the order parameter fluctuations are not totally ignored and the ”photon mass” $\rho(\psi)$ generated by the effect of the Higgs field $\psi$ on the vector potential $A$ is not an equilibrium quantity. The special feature of the present method is that the uniform fluctuation $\delta \psi$ of the uniform ”field” $\psi$ interacts with the magnetic fluctuations and this interaction does ensure the fluctuation mechanism of the FOT transition discussed in the present and all preceding works. The WFOT in higher spatial dimensionalities and the FOT in our case are caused by a high-order fluctuation interaction $[(\delta \psi)^2(\delta A_j)^2]$ between the order parameter and the magnetic field fluctuations rather than the magnetic fluctuations alone.

(3) The theoretical arguments in favour of the FOT within the approximation $\psi = \text{const}$ below the equilibrium transition point are not precisely the same as those obtained by various renormalization group investigations in one loop [1, 7, 13] and higher-order [20, 21, 22] approximations in the loop expansion, where another type of singularities of the perturbation series for the GL free energy are relevant. Within the present approximation for $\psi$ the divergent terms, which are usually relevant for the renormalization group studies and are present in all diagrammes in Fig. 1, have been summed up to a finite sum. Therefore, the arguments in favour (or against) a WFOT within the approximation of uniform scalar field $\psi$ cannot be used to justify or reject renormalization group predictions above the equilibrium transition point.
Acknowledgments:

The authors thank the OSI (Vienna and its extension in Sofia) for a research grant. DIU thanks the hospitality of the Johannes Kepler Universität (Linz) and the Max-Planck-Institut für Komplexer Systeme (Dresden).
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Figure captions

Fig. 1. One loop logarithmic series of diagrammes: the black squares denote the vertex part of the $A_j^2$– term, and the solid lines denote the correlation function $< |A_j(k)|^2 >$. 

Fig. 2. The shape of the effective free energy $f$ for $q = 10^{-4}$: line (1) corresponds to $r = -9.3 \times 10^{-4}$, (2) – to $r = -9.4 \times 10^{-4}$, (3) – to $r = -9.515 \times 10^{-4}$, and (4) – to $r = -9.7 \times 10^{-4}$.

Fig. 3. The order parameter profile for $q = 10^{-4}$. The black squares indicate the stable superconducting states and the white squares indicate metastable superconducting states above the equilibrium transition point.
