NEW FEATURES OF THE PHASE TRANSITION TO
SUPERCONDUCTING STATE IN THIN FILMS

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Abstract

The Halperin-Lubensky-Ma (HLM) effect of a fluctuation-induced change of the order of phase transition in thin films of type I superconductors with relatively small Ginzburg-Landau number $\kappa$ is considered. Numerical data for the free energy, the order parameter jump, the latent heat, and the specific heat of W, Al and In are presented to reveal the influence of film thickness and material parameters on the properties of the phase transition. We demonstrate for the first time that in contrast to the usual notion the HLM effect occurs in the most distinct way in superconducting films with high critical magnetic field $H_{c0}$ rather than in materials with small $\kappa$. The possibility for an experimental observation of the fluctuation change of the order of superconducting phase transition in superconducting films is discussed.

1. Introduction

Our paper is intended to clarify the best conditions for an experimental observation of the Halperin-Lubensky-Ma effect (HLM) \[1, 2\] of a fluctuation-induced first-order phase transition from normal to Meissner phase in a zero external magnetic field for thin films of type I superconductors \[3, 4, 5\]. For this purpose we present new theoretical results about the thermodynamics of the phase transition from normal to superconducting state in a zero external magnetic field.

The HLM effect is predicted theoretically \[1\] to occur in pure \[2, 4, 6\] and disordered \[7, 8, 9\] bulk, (three-dimensional - 3D), and 2D \[10\] superconductors, as well as in quasi-2D superconducting films \[3, 5\] but up to now it has not been observed in experiments. The calculated effect is very small in 3D superconductors and is not possible to be detected even
for a high purity of the sample and perfection of the crystal lattice [1, 2, 4]. Recently, it
has been shown [3, 5] that in thin (quasi-2D) films the HLM effect is much stronger than
in 3D samples and could be observed by available experimental techniques if the type of
superconductor and film thickness are properly chosen for the experiments; for a review,
see also, Refs. [11, 12]. This result gives an opportunity to search for the effect in suitable
superconducting films.

The HLM effect appears as a result of the interaction between the superconducting order
parameter $\psi(x)$ and the vector potential $\vec{A}(x)$ of the magnetic induction in the Ginzburg-
Landau (GL) free energy of a superconductor. According to the theoretical paradigm intro-
duced for the first time in the scalar electrodynamics by Coleman and Weinberg (CW) [13],
this effect should occur in all physical systems described by Abelian-Higgs models where
a scalar gauge field (like $\psi$ in superconductors) interacts in a gauge invariant way with
another vector gauge field (like the vector potential $\vec{A}$ in superconductors). In addition
to the mentioned examples of superconductors and scalar electrodynamics, the same type
of interaction plays an important role in the nematic-smectic A phase transition in liquid
crystals [14, 15, 16] and phase transitions in the early universe [17]. HLM effect may be
also relevant to quantum phase transitions in superconductors [18, 19, 20] and itinerant
ferromagnets [21].

On the other side, there are certain theoretical investigations, based on Monte Carlo simula-
tions [22] and the so-called “dual model” [23] which do not confirm the fluctuation-change
of the order of the phase transition (see also Refs. [12]). That is why, extensive experi-
ments intended to verify the existence of the effect were made in liquid crystals; see, e.g.,
Refs. [12]. But in liquid crystals the weakly-first order phase transition predicted by CW
and HLM can be obscured by similar effects due to the strong crystal anisotropy, while the
recent result [3] about the considerable enhancement of HLM effect in suitable supercon-
ducting films can be used for more reliable experiments. For this aim we need to find the
best material parameters and the most suitable film thickness, having in mind some purely
experimental problems that may appear.

Recently, we partly solved the problem for Al films [5] nevertheless additional theoretical
investigations should be done. In this paper we shall present some new theoretical predic-
tions for Al films as well as new numerical data for thin films of W and In. The choice
of this element superconductors is made for their relatively small GL number $\kappa = (\lambda/\xi)$,
which allows a more distinct appearance of the HLM effect in both bulk and thin film
superconductors [1, 3, 5]: here $\lambda$ is the London penetration depth and $\xi$ is the coherence
length [24].

We focus our attention on numerical data for the behavior of the free energy and directly
measurable thermodynamic quantities like the order parameter jump, the latent heat, and
the specific heat. A surprising result of our analysis of the data for W, Al, and In is that
the HLM effect in thin films is stronger in case of relatively high zero-temperature critical
magnetic field $H_{c0}$ rather than for relatively small GL number $\kappa$, as claimed in preceding
Our investigation is based on the theoretical results from preceding papers \[1, 2, 3, 4, 5\]. In Sec. 2 we shall outline the theoretical framework of our study. In Sec. 3 we present our analysis of thin films of tungsten (W), aluminium (Al), and indium (In) and a discussion of the results with a special emphasis on their application to experiments. In Sec. 4 we summarize our findings.

2. Theoretical basis

Our investigation is based on the Ginzburg-Landau free energy \[24\] of a D-dimensional superconductor with volume \( V = (L_1 \ldots L_D) \) given by

\[
F = \int d^D x \left[ a |\psi|^2 + \frac{b}{2} |\psi|^4 + \frac{\hbar^2}{4m} \left( \nabla - \frac{2i e}{\hbar c} \vec{A} \right) \psi \right]^2 + \frac{1}{16\pi} \sum_{i,j=1}^{3} \left( \frac{\partial A_i}{\partial x_j} - \frac{\partial A_j}{\partial x_i} \right)^2 , \tag{1}
\]

where \( a = \alpha_0 (T - T_{c0}) \) and \( b > 0 \), are the Landau parameters, \( e = |e| \) is the electron charge, \( \psi(\vec{x}) \) is the order parameter, and \( \vec{A}(\vec{x}) \) is the vector potential of the magnetic field.

The critical temperature \( T_{c0} \) corresponds to the second order phase transition which occurs in a zero external magnetic field when the fluctuations, \( \delta \varphi(\vec{x}) \) and \( \delta \vec{A}(\vec{x}) \), of both fields \( \psi(\vec{x}) \) and \( \vec{A}(\vec{x}) \) are neglected. Usually, this case is considered in the low-temperature superconductors, where the effect of the superconducting fluctuations \( \delta \varphi(\vec{x}) \) on the thermodynamics is very small and practically uninteresting, and the same has been supposed for the magnetic fluctuations \( \delta \vec{A}(\vec{x}) \) before the appearance of HLM paper \[1\]. In our study the superconducting fluctuations are ignored as negligibly small which is a suitable approximation in type I superconductors where \( \lambda \ll \xi \). But we take into account the magnetic fluctuations to a full extent. Then the normal-to-superconducting phase transition in a zero external magnetic field turns out of first order at an equilibrium phase transition temperature \( T_{eq} \) that is different from \( T_{c0} \). So, our task will be to point the type of superconductors, where this picture may be valid and investigate the properties of the first order phase transition.

We shall follow the theoretical approach described in details in preceding papers \[1, 2, 3, 5\], where an effective free energy \( F_{\text{eff}}(\psi) \) of the type I superconducting film was obtained \[3\]. There we also neglected the superconducting fluctuations and did the calculation of the mean-field value of the uniform (\( \vec{x} \)-independent) superconducting order parameter \( \psi \) in a self-consistent way after taking into account the magnetic fluctuations through an exact integration out of the field \( \vec{A}(\vec{x}) \) in the partition function of the superconductor. As the external magnetic field is equal to zero the regular part \( \vec{A}_0 = (\vec{A} - \delta \vec{A}) \) of \( \vec{A} \) related to it can be set equal to zero, too. Then \( \vec{A} = \delta \vec{A} \), therefore, we consider the net effect of the magnetic fluctuations.

Having in mind that in type I superconductors a stable vortex phase cannot occur, we again can assume that the order parameter \( \psi \) that describes the uniform Meissner phase in the bulk of the superconducting film is \( \vec{x} \)-independent. Our investigation is based on the quasi-macroscopic GL theory so we must consider films of thickness \( L_0 \gg a_0 \), where
\( \alpha_0 \) is the lattice constant. In such films the surface energy can be ignored and one can use periodic boundary conditions without a substantial departure from the real situation. In this way, the surface effects as a source of a spatial dependence of the order parameter \( \psi \) are also eliminated.

Following \[11, 2, 3, 4, 5\] we present the effective free energy density \( f(\psi) = F_{\text{eff}}(\psi)/V \) of a 3D superconducting slab of volume \( V = (L_1 L_2 L_0) \) and thickness \( L_0 \) in the form:

\[
f(\varphi) = \frac{H_c^2}{8\pi} \left\{ 2t_0 \varphi^2 + \varphi^4 + C(1 + t_0) \left[ (1 + \mu \varphi^2) \ln (1 + \mu \varphi^2) - \mu \varphi^2 \ln (\mu \varphi^2) \right] \right\},
\]

where

\[
C = \frac{2\pi^2 k_B T_{c0}}{L_0 \xi_0^2 H_c^2}.
\]

Here \( \varphi = (|\psi|/|\psi_0|) \) is the dimensionless order parameter defined with the help of the zero-temperature value \( |\psi_0| = |\psi(T = 0)| = (\alpha_0 T_{c0}/b)^{1/2} \) of \( \psi \), \( t_0 = (T - T_{c0})/T_{c0} \), \( \mu = (\xi_0/\pi \lambda_0)^2 \) is given by the zero-temperature value \( \xi_0 = (\hbar^2/4m_0 T_{c0})^{1/2} \) of \( \xi \) and \( \lambda_0 = (b/\rho_0 \alpha_0 T_{c0})^{1/2} \) is the zero-temperature penetration depth; \( \rho_0 = 8\pi e^2/m c^2 \). We also use the notations: \( \lambda(T) = \lambda_0/|t_0|^{1/2} \) and \( \xi(T) = \xi/|t_0|^{1/2} \). The critical magnetic field at \( T = 0 \) is given by \[24\] \( H_{c0} = \alpha_0 T_{c0}(4\pi/b)^{1/2} \). The relations of \( H_{c0} \) and \( \xi_0 \) with \( b \) and \( \alpha_0 \), respectively, can be used together with the experimental data for \( H_{c0} \) and \( \xi_0 \) in concrete superconducting substances in order to calculate the parameters \( b \) and \( \alpha_0 \).

The equilibrium order parameter \( \varphi_0 > 0 \) corresponding to the Meissner phase can be easily obtained from the equation \( \partial f(\varphi)/\partial \varphi = 0 \) and Eq. \[2\]:

\[
t_0 + \varphi_0^2 + \frac{C \mu(1 + t_0)}{2} \left[ \ln \left( 1 + \frac{1}{\mu \varphi_0^2} \right) - \frac{1}{1 + \mu \varphi_0^2} \right] = 0.
\]

The logarithmic divergence in Eq. \[4\] has no chance to occur because \( \varphi_0 \) is always positive and does not tend to zero.

We shall use the notations from Ref. \[5\] for the entropy jump \( \delta s \) and the specific heat jump \( \delta C \) at the equilibrium phase transition point \( T_{\text{eq}} \) of first order corresponding to a zero external magnetic field. Here we shall give the previously calculated results for the leading terms in these quantities (terms of higher order are neglected as small), namely,

\[
\delta s = -\frac{H_c^2}{4\pi T_{c0}} \varphi_{\text{eq}}^2,
\]

and

\[
\delta C = \frac{H_c^2}{4\pi T_{c0}}.
\]

The latent heat of the phase transition is given by \( Q = T_{\text{eq}} \delta s \) and Eq. \[4\]. Since the temperatures \( T_{\text{eq}} \) and \( T_{c0} \) have very close values, the difference between the values of \( Q \), \( \delta s \), and \( \delta C \) at \( T_{c0} \) and \( T_{\text{eq}} \), respectively, can also be ignored, for example, \( |\delta C(T_{\text{eq}}) - \delta C(T_{c0})|/\delta C(T_{c0}) \ll 1 \) and we can use either \( \delta C(T_{c0}) \) or \( \delta C(T_{\text{eq}}) \) \[5\]. Here the jumps \( \delta s \),
\( \delta C \), and \( Q \) are all taken at the equilibrium phase transition value \( T_{eq} \) but we shall not supply them with the subscript “eq” as we do for other quantities.

Eqs. (2) - (6) are valid for thin films \( (a_0 \ll L_0 \sim \xi_0) \) in a zero external magnetic field \( \vec{H} \) and for negligibly small \( \psi \)-fluctuations which means that they are applicable for low-temperature nonmagnetic superconductors \((T_{c0} < 20K)\). Because in experiments the external magnetic field cannot be completely eliminated, vortex states may occur for \( H = |\vec{H}| > 0 \) below \( T_c = T_c(H) \leq T_{c0} \) in type II superconducting films and this will obscure the HLM effect. Note, that the magnetic field \( H \) generates additional entropy jump at the phase transition point \( T_c(H) \) and this effect can hardly be separated from the entropy jump (5) caused by the magnetic fluctuations in the close vicinity of \( T_{c0} \). Therefore, in experiments intended to a search of the HLM effect we must choose type I superconductors. The second important point is connected with the value of the square \( \varphi_{eq}^2 \) which is proportional to the superconducting current \( (j_s \sim |\psi|_{eq}^2 \sim \varphi_{eq}^2) \) and to the equilibrium jumps \( \delta s \) and \( \delta Q \). In 3D superconductors the ratio \((Q/\delta C)\) depends on \( H_{c0}^2 \xi_0 \sim \epsilon_c \kappa^{-6} \), where \( \epsilon_c \sim 10^{-16} \) denotes the extremely small Ginzburg-Levanyuk critical region \([11]\) of low-temperature superconductors \([1]\). Therefore, the latent heat in these 3D superconductors can hardly be observed in experiments. But in thin films the substantial dependence of the entropy \( \delta s \) and the specific heat \( \delta C \) is on the critical magnetic field \( H_{c0}^2 \), as shown by Eqs. (5) - (6) and the analysis in Sec. 3.

The equations (2) and (4) corresponding to quasi-2D films are quite different from the respective equations \([1, 2]\) for bulk (3D-) superconductors but it is easily seen that the relatively large values of the order parameter jump \( \varphi^2 \) in thin films again correspond to relatively small values of the GL parameter \( \kappa \). That is why we consider element superconductors with small values of \( \kappa \) and study the effect of this parameter, the critical magnetic field \( H_{c0} \) and the film thickness \( L_0 \) on the properties of the fluctuation-induced first order phase transition.

Theoretical results we have used in this Section for quasi-2D superconducting films are consistent with the theory \([25]\) of 2D-3D crossover phenomena near phase transition points and the 2D-3D crossover theory \([26, 27]\) of the HLM effect; see also Ref. \([28]\).

### 3. Results and discussion

We use experimental data for \( T_{c0}, H_{c0}, \xi_0 \) and \( \kappa \) for W, Al, and In published in Ref. \([29]\) (see Table 1). In some cases the GL parameter \( \kappa \) can be calculated with the help of the relation \( \kappa = (\lambda_0/\xi_0) \) and the available data for \( \xi_0 \) and \( \lambda_0 \). In other cases it is more convenient to use the following representation of the zero-temperature penetration depth:

\[
\lambda_0 = \frac{\hbar c}{2\sqrt{2}eH_{c0}\xi_0} .
\]  

The value of \(|\psi_0|\) in Table 1 is found from

\[
|\psi_0| = \left( \frac{m}{\pi \hbar^2} \right)^{1/2} \xi_0 H_{c0} .
\]  

5
Eqs. (7) and (8) are obtained from the formulae given after Eq. (3). Besides we calculate the parameter $\tilde{C} = CL_0$ with the help of Eq. (3) and the data in Table 1.

Note, that the experimental data vary within 5-10% depending on the experimental technique used in measurements. Moreover, these data correspond to bulk samples and may differ within 10-20% from those for very thin films ($L_0 < 10^{-2}\mu$m). However, these variations in the experimental data do not essentially affect our results.

Table 1. Values of $T_0$, $H_0$, $\xi_0$, $\kappa$, and $|\psi_0|$ for W, Al, In.

| substance | $T_0$ (K) | $H_0$ (Oe) | $\xi_0$ (\mu m) | $\kappa$ | $|\psi_0| \times 10^{-11}$ |
|-----------|-----------|-----------|-----------------|---------|------------------|
| W         | 0.015     | 1.15      | 37              | 0.001   | 0.69              |
| Al        | 1.19      | 99.00     | 1.16            | 0.010   | 2.55              |
| In        | 3.40      | 281.5     | 0.44            | 0.145   | 2.0               |

Table 2. Values of $t_{eq}$, $\varphi_{eq}$, and $Q$ (erg/cm$^3$) for films of W, Al, and In with different thicknesses $L_0$ (\mu m).

| $L_0$ | $t_{eq}$ | $\varphi_{eq}$ | $Q$   | $t_{eq}$ | $\varphi_{eq}$ | $Q$   | $t_{eq}$ | $\varphi_{eq}$ | $Q$   |
|-------|---------|-----------------|------|---------|-----------------|------|---------|-----------------|------|
| 0.05  | -0.00230| 0.041           | -1.95| -0.00167| 0.025           | -3.94| -0.00174| 0.039           | -1.6 \times 10^{-4} |
| 0.1   | -0.00147| 0.032           | -0.80| -0.00094| 0.017           | -1.82| -0.00118| 0.032           | -1.1 \times 10^{-4} |
| 0.3   | -0.00070| 0.023           | -0.41| -0.00037| 0.010           | -0.63| -0.00064| 0.023           | -5.6 \times 10^{-5} |
| 0.5   | -0.00048| 0.016           | -0.20| -0.00029| 0.008           | -0.40| -0.00048| 0.020           | -4.1 \times 10^{-5} |
| 1     | -0.00029| 0.012           | -0.11| -0.00013| 0.006           | -0.23| -0.00032| 0.016           | -2.7 \times 10^{-5} |
| 2     | -0.00017| 0.009           | -0.06| -0.00008| 0.004           | -0.10| -0.00021| 0.013           | -1.8 \times 10^{-5} |

The order parameter profile for Al films of different thicknesses is shown in Fig. 1. It is readily seen that the behavior of the function $\varphi_0(t_0)$ corresponds to a well established phase transition of first order. The vertical dashed lines in Fig. 1 indicate the respective values of $t_{eq} = t_0(T_{eq})$, at which the equilibrium phase transition occurs as well as the equilibrium jump $\varphi_0(T_{eq}) = \varphi_{eq}$ for different thicknesses of the film. The parts of the $\varphi_0(t_0)$-curves which extend up to $t_0 > t_{eq}$ describe the metastable (overheated) Meissner states which can appear under certain experimental circumstances (see in Fig. 1 the parts of the curves on the r.h.s. of the dashed lines). The value of $\varphi_{eq}$ and the metastable region decrease with the increase of the film thickness, which shows that the first order of the phase transition is better pronounced in thinner films and that confirms a conclusion in Ref. [20].

These results are confirmed by the behavior of the free energy as a function of $t_0$. We used Eqs. (2) and (4) for the calculation of the equilibrium free energy $f[\varphi_0(t_0)]$. The free energy for Al films with different thicknesses is shown in Fig. 2. The equilibrium points $T_{eq}$ of the phase transition correspond to the intersection of the $f(\varphi_0)$-curves with the $t_0$-axis. It is obvious from Fig. 2 that the temperature domain of overheated Meissner states decreases with the increase of the thickness $L_0$.

The shape of the equilibrium order parameter $\varphi_0$ in a broad vicinity of the equilibrium
Figure 1: Order parameter profile $\varphi(t_0)$ of Al films of different thicknesses: $L_0 = 0.05$ $\mu$m (‘+’-line), $L_0 = 0.1$ $\mu$m (○), and $L_0 = 0.3$ $\mu$m (·).

Figure 2: The free energy $f(t_0)$ for Al films of thickness: $L_0 = 0.05$ $\mu$m (‘+’-line), $L_0 = 0.1$ $\mu$m (○), $L_0 = 0.3$ $\mu$m (·).
phase transition of thin films \((L_0 = 0.05 \mu m)\) of W, Al, and In was found from Eq. (4). The result is shown in Fig. 3. The vertical dashed lines in Fig. 3 again indicate the respective values of \(t_{eq} = t_0(T_{eq})\), at which the equilibrium phase transition occurs as well as the equilibrium jump \(\varphi_0(T_{eq}) = \varphi_{eq}\) in the different superconductors.

The order parameter jump at the phase transition point of In (the In curve is marked by points in Fig. 3) is relatively smaller than for W, and Al, where the GL parameter has much lower values. The same is valid for the metastability domains; see the parts of the curves in Fig. 3 on the left of the vertical dashed lines. It is obvious from Fig. 3 and Table 2 that the equilibrium jump of the reduced order parameter \(\varphi_{eq}\) of W has a slightly smaller value than that of Al although the GL number \(\kappa\) for W has a ten times lower value compared with \(\kappa\) of Al. Note, that in Fig 3 we show the jump of \(\varphi_{eq}\), but the important quantity is \(|\psi|_{eq} = |\psi_0|\varphi_{eq}\). Using the data for \(L_0 = 0.05 \mu m\) from Tables 1 and 2 we find for \(|\psi|_{eq}\) the following values: \(0.1 \times 10^{11}\) for Al, \(0.05 \times 10^{11}\) for In, and \(0.02 \times 10^{11}\) for W. This result shows that the value of the critical field \(H_{c0}\) is also important and should be taken into account together with the smallness of GL number when the maximal values of the order parameter jump are looked for. Thus the value of the order parameter jump at the fluctuation-induced phase transition is maximal provided small values of the GL parameter \(\kappa\) are combined with relatively large values of the critical field \(H_{c0}\). In our case Al has the optimal values of these two parameters.

The shift of the phase transition temperature \(t_{eq} = |(T_{eq} - T_{c0})|/T_{c0}\), the reduced value \(\varphi_{eq}\) of the equilibrium order parameter jump \(|\psi|_{eq}\), and the latent heat \(Q\) of the equilib-
rium transition are given for films of different thicknesses and substances in Table 2. The thicknesses are chosen so as to ensure the validity of the theory used in our analysis and to satisfy other important requirements presented in Sec. 4. The data in Table 2 show that the shift of the phase transition temperature is very small and can be neglected in all calculations and experiments based on them. The values for \( \varphi_{eq} \) for different \( L_0 \) and those for \( |\psi_0| \) given in Table 1 confirm the conclusion which we have made for films of Al, In, and W with \( L_0 = 0.05 \mu m \). The latent heat \( Q \) has maximal values for In, where the critical field is the highest for the considered materials.

4. Conclusion

In contrast to our initial expectations that films made of superconductors with extremely small GL parameter \( \kappa \) such as Al and, in particular, W will be the best candidates for an experimental search of the HLM effect, our careful analysis definitely gives somewhat different answer. The Al films still remain a good candidate for transport experiments through which the jump of the order parameter at the phase transition point could be measured but surprisingly the W films turn out inconvenient for the same reason because of their very low critical field \( H_{c0} \). Although In has ten times higher GL number \( \kappa \) than Al, the In films can be used on an equal footing with the Al films in experiments intended to prove the order parameter jump. Here the choice of one of these materials may depend on other features of experimental convenience. As far as caloric experiments are concerned, the In films seem the best candidate for their high latent heat.

We have presented the theoretical justification and predictions intended to support experiments on the observation of magnetic fluctuations and HLM effect near the normal-to-superconducting transition in a zero external magnetic field. Besides, we have demonstrated for the first time that the experiments can be most successfully performed in type I superconductors with relatively high critical magnetic fields.

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