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New features of the phase transition to the 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 κ 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 κ . The possibility of an experimental observation of the fluctuation change of the order of the superconducting phase transition in superconducting films is discussed.

1. Introduction

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

The HLM effect is predicted theoretically [1] to occur in pure [2, 4, 6] and disordered [7–9] 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 detect even for a high purity sample with a perfect crystal lattice [1, 2, 4]. Recently, it has been shown [3, 5]

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that in thin (quasi-2D) films the HLM effect is much stronger than in 3D samples and can be observed by available experimental techniques if the type of superconductor and film thickness are properly chosen for the experiment; for a review, see also [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 $\psi(\vec{x})$ and the vector potential $\vec{A}(\vec{x})$ of the magnetic induction in the Ginzburg–Landau (GL) free energy of a superconductor. According to the theoretical paradigm introduced 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 ψ 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 [14–16] and phase transitions in the early universe [17]. The HLM effect may also be relevant to quantum phase transitions in superconductors [18–20] and itinerant ferromagnets [21].

On the other hand, there are certain theoretical investigations, based on Monte Carlo simulations [22] and the so-called ‘dual model’ [23], which do not confirm the fluctuation change of the order of the phase transition (see also [12]). That is why extensive experiments intended to verify the existence of the effect were performed in liquid crystals; see, e.g., [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] on the considerable enhancement of the HLM effect in suitable superconducting films can be used for more reliable experiments. To achieve this aim we need to find the best material parameters and the most suitable film thickness, bearing 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 predictions for Al films as well as new numerical data for thin films of W and In. The choice of these superconductor elements 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, λ is the London penetration depth and ξ is the coherence length [24].

We focus our attention on numerical data for the behaviour 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 the case of relatively high zero-temperature critical magnetic field H_{c0} rather than for relatively small GL number κ , as claimed in preceding papers [1–5].

The investigation is based on the theoretical results from preceding papers [1, 3–5]. In section 2 we shall outline the theoretical framework of our study. In section 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 section 4 we summarize our findings.

2. Theoretical basis

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

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

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 was supposed for the magnetic fluctuations $\delta\vec{A}(\vec{x})$ before the appearance of the 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 the full extent. Then the normal-to-superconducting phase transition in a zero external magnetic field turns out to be of first order at an equilibrium phase transition temperature T_{eq} that is different from T_{c0} . So, our task will be to point to the type of superconductor where this picture may be valid, and investigate the properties of the first order phase transition.

We shall follow the theoretical approach described in detail in preceding papers [1–3, 5], where an effective free energy $F_{\text{eff}}(\psi)$ of the type I superconducting film was obtained [5]. There we also neglected the superconducting fluctuations and performed the calculation of the mean-field value of the uniform (\vec{x} -independent) superconducting order parameter ψ 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.

The effective free energy $F_{\text{eff}}(\psi) = -k_B T \ln Z(\psi)$ can be obtained from the solution of the partition integral $Z(\psi)$ for the vector potential fluctuations $\vec{A}(\vec{x})$. The partition function $Z(\psi)$ is defined with the help of the statistical weight $\exp[-F(\psi, \vec{A})/k_B T]$, where $F(\psi, \vec{A})$ is the GL free energy (1) and the spatial variations of the field $\psi(\vec{x})$ are neglected. So, a functional Gaussian integral $Z(\psi)$ has to be solved over all configurations of the field $\vec{A}(\vec{x})$ that obey the Coulomb gauge $\text{div}\vec{A}(\vec{x}) = 0$. This can be done exactly either by a direct integration or by a loop-wise expansion; see [3], which can be summed up to the logarithmic function shown below.

Bearing in mind that in type I superconductors a stable vortex phase cannot occur, we can again assume that the order parameter ψ 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 a_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 ψ are also eliminated.

Following [1–5] 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

$$f(\varphi) = \frac{H_{c0}^2}{8\pi} \{2t_0\varphi^2 + \varphi^4 + C(1 + t_0)[(1 + \mu\varphi^2) \ln(1 + \mu\varphi^2) - \mu\varphi^2 \ln(\mu\varphi^2)]\}, \quad (2)$$

where

$$C = \frac{2\pi^2 k_B T_{c0}}{L_0 \xi_0^2 H_{c0}^2}. \quad (3)$$

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 $|\psi|$, $t_0 = (T - T_{c0})/T_{c0}$ will be called a reduced temperature difference, the parameter $\mu = (\xi_0/\pi\lambda_0)^2$ is given by the zero-temperature value $\xi_0 = (\hbar^2/4m\alpha_0 T_{c0})^{1/2}$ of ξ and $\lambda_0 = (b/\rho_0\alpha_0 T_{c0})^{1/2}$ is the zero-temperature

penetration depth ($\rho_0 = 8\pi e^2/mc^2$). We also use the notation $\lambda(T) = \lambda_0/|t_0|^{1/2}$ and $\xi(T) = \xi_0/|t_0|^{1/2}$. 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 ξ_0 with b and α_0 , respectively, can be used together with the experimental data for H_{c0} and ξ_0 in concrete superconducting substances in order to calculate the parameters b and α_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 equation (2):

$$t_0 + \varphi_0^2 + \frac{C\mu(1+t_0)}{2} \ln\left(1 + \frac{1}{\mu\varphi_0^2}\right) = 0. \quad (4)$$

The logarithmic divergence in equation (4) has no chance to occur because φ_0 is always positive and does not tend to zero.

We shall use the notations from [5] for the entropy jump δs and the specific heat jump δC at the equilibrium first order phase transition point T_{eq} 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,

$$\delta s = -\frac{H_{c0}^2}{4\pi T_{c0}} \varphi_{\text{eq}}^2, \quad (5)$$

and

$$\delta C = \frac{H_{c0}^2}{4\pi T_{c0}}. \quad (6)$$

The latent heat of the phase transition [11] is given by $Q = -T_{\text{eq}}\delta s$ and equation (4). Since the temperatures T_{eq} and T_{c0} have very close values, the difference between the values of Q , δs and δC at T_{c0} and T_{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 δs , δC and Q are all taken at the equilibrium phase transition value T_{eq} but we shall not give them the subscript 'eq' as we do for other quantities.

Equations (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 ψ -fluctuations which means that they are applicable for low-temperature nonmagnetic superconductors ($T_{c0} < 20$ K). 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 an 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 for the HLM effect we must choose type I superconductors. The second important point is connected with the value of the square φ_{eq}^2 which is proportional to the superconducting current ($j_s \sim |\psi|_{\text{eq}}^2 \sim \varphi_{\text{eq}}^2$ [24]) and to the equilibrium jumps δs and δQ . 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 δs and the specific heat δC is on the critical magnetic field H_{c0}^2 , as shown by equations (5), (6) and the analysis in section 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 φ^2 in thin films again correspond to relatively small values of the GL parameter κ . That is why we consider element superconductors with small

Table 1. Values of T_{c0} , H_{c0} , ξ_0 , κ and $|\psi_0|$ for W, Al, In.

Substance	T_{c0} (K)	H_{c0} (Oe)	ξ_0 (μm)	κ	$ \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

values of κ 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 [28].

3. Results and discussion

We use experimental data for T_{c0} , H_{c0} , ξ_0 and κ for W, Al and In published in [29] (see table 1). In some cases the GL parameter κ can be calculated with the help of the relation $\kappa = (\lambda_0/\xi_0)$ and the available data for ξ_0 and λ_0 . In other cases it is more convenient to use the following representation of the zero-temperature penetration depth:

$$\lambda_0 = \frac{\hbar c}{2\sqrt{2}eH_{c0}\xi_0}. \quad (7)$$

The value of $|\psi_0|$ in table 1 is found from

$$|\psi_0| = \left(\frac{m}{\pi\hbar^2}\right)^{1/2} \xi_0 H_{c0}. \quad (8)$$

Equations (7) and (8) are obtained from the formulae given after equation (3). We also calculate the parameter $\tilde{C} = CL_0$ with the help of equation (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 ($L_0 < 10^{-2} \mu\text{m}$). However, these variations in the experimental data do not essentially affect our results.

The order parameter dependence on the reduced temperature difference t_0 is shown in figure 1 for Al films of different thicknesses. It is readily seen that the behaviour of the function $\varphi_0(t_0)$ corresponds to a well established phase transition of first order. The vertical dashed lines in figure 1 indicate the respective values of $t_{\text{eq}} = t_0(T_{\text{eq}})$, at which the equilibrium phase transition occurs as well as the equilibrium jump $\varphi_0(T_{\text{eq}}) = \varphi_{\text{eq}}$ for different thicknesses of the film. The parts of the $\varphi_0(t_0)$ -curves which extend up to $t_0 > t_{\text{eq}}$ describe the metastable (overheated) Meissner states which can appear under certain experimental circumstances (see in figure 1 the parts of the curves on the rhs of the dashed lines). The value of φ_{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 [20].

These results are confirmed by the behaviour of the free energy as a function of t_0 . We used equations (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 figure 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 figure 2 that the temperature domain of overheated Meissner states decreases with the increase of the thickness L_0 .

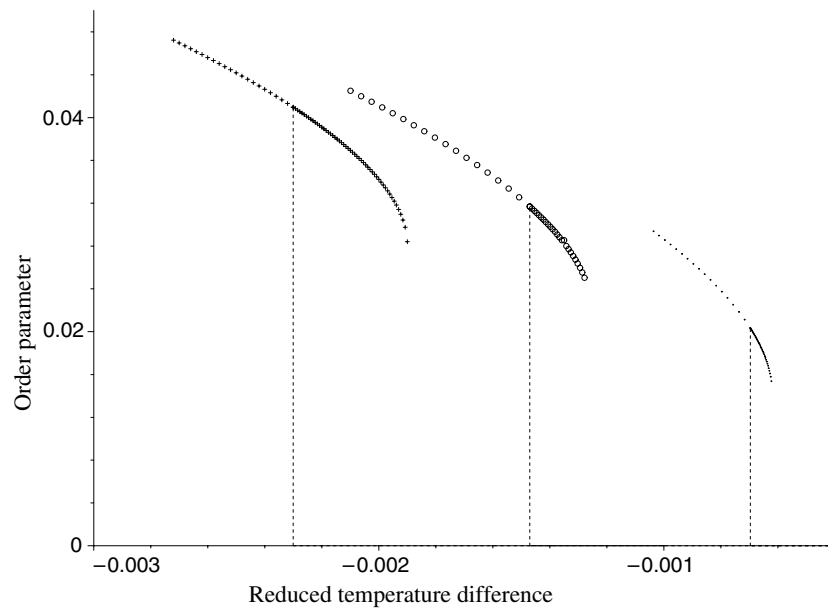


Figure 1. Order parameter profile $\varphi(t_0)$ of Al films of different thicknesses: $L_0 = 0.05 \mu\text{m}$ (+), $L_0 = 0.1 \mu\text{m}$ (O) and $L_0 = 0.3 \mu\text{m}$ (·).

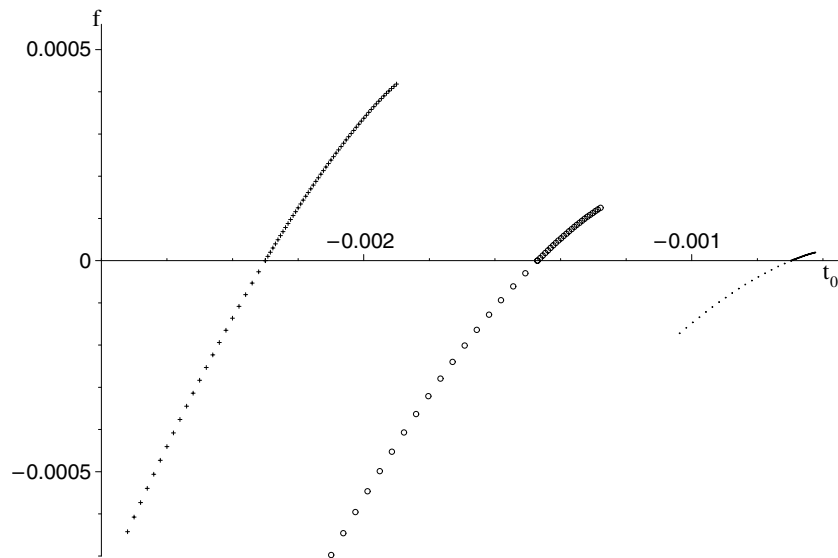


Figure 2. The free energy $f(t_0)$ for Al films of thickness $L_0 = 0.05 \mu\text{m}$ (+), $L_0 = 0.1 \mu\text{m}$ (O) and $L_0 = 0.3 \mu\text{m}$ (·).

The shape of the equilibrium order parameter $\varphi_0(t_0)$ in a broad vicinity of the equilibrium phase transition of thin films ($L_0 = 0.05 \mu\text{m}$) of W, Al and In was found from equation (4). The result is shown in figure 3. The vertical dashed lines in figure 3 again indicate the respective values of $t_{\text{eq}} = t_0(T_{\text{eq}})$ at which the equilibrium phase transition occurs as well as the equilibrium jump $\varphi_0(T_{\text{eq}}) = \varphi_{\text{eq}}$ in the different superconductors.

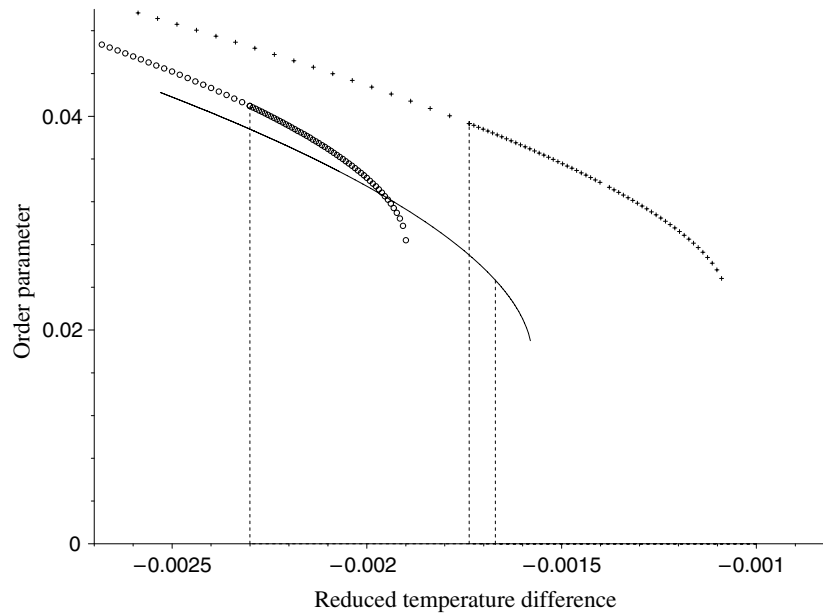


Figure 3. Order parameter profile $\varphi(t_0)$ of films of thickness $L_0 = 0.05 \mu\text{m}$: W (+), Al (O) and In (·).

The order parameter jump at the phase transition point of In (the In curve is marked by points in figure 3) is 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 figure 3 on the left of the vertical dashed lines. It is obvious from figure 3 and table 2 that the equilibrium jump of the reduced order parameter φ_{eq} of W has a slightly smaller value than that of Al although the GL number κ for W has a ten times lower value compared with κ of Al. Note that in figure 3 we show the jump of φ_{eq} , but the important quantity is $|\psi|_{\text{eq}} = |\psi_0|\varphi_{\text{eq}}$. Using the data for $L_0 = 0.05 \mu\text{m}$ from tables 1 and 2 we find for $|\psi|_{\text{eq}}$ the following values: 0.1×10^{11} for Al, 0.05×10^{11} for In and 0.02×10^{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 the 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 κ 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 importance of the zero-temperature critical magnetic field H_{c0} for the enhancement of the jumps of the certain thermodynamic quantities at the equilibrium phase transition point T_{eq} becomes obvious from equations (5), (6) and (8). Equation (8) shows that the order parameter jump $|\psi|_{\text{eq}} = |\psi_0|\varphi_{\text{eq}}$ is large for large values of H_{c0} and ξ_0 . This is consistent with the requirement for relatively small values of the GL parameter κ as shown by equation (7). Therefore, the unmeasurable ratio $Q/\delta C$ discussed in [1] does not depend on the value of the critical field H_{c0} but the quantities Q and δC themselves as well as the order parameter jump $|\psi|_{\text{eq}}$ depend essentially on H_{c0} . The values of the reduced order parameter jump φ_{eq} for films of Al, In and W of the same thickness have the same order of magnitude while the respective order parameter jump $|\psi|_{\text{eq}}$ is one order of magnitude higher for Al than for W, as shown above. The effect of the critical magnetic field H_{c0} on the latent heat Q is, however, much stronger

Table 2. Values of t_{eq} , φ_{eq} and Q (erg cm⁻³) for films of W, Al and In with different thicknesses L_0 (μm).

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

and, as is seen from table 2, the latent heat Q in W films is very small and can be neglected while in Al and In films it reaches values which could be measured in suitable experiments. This is so because the latent heat is proportional to the difference [$H_{c0}^2/8\pi \sim b|\psi_0|^4$] between the energies of the ground state (superconducting phase at $T = 0$) and the normal state. It is consistent with the fact that the fluctuation contribution to the free energy, i.e., the C -term in the rhs of equation (2), is generated by the term of type $|\psi|^2 \int d^D x \vec{A}^2(\vec{x})$ in the GL free energy (1). At $T = 0$ this free energy term is also proportional to the mentioned difference between the free energies of the ground and normal states.

The shift of the phase transition temperature $t_{\text{eq}} = |(T_{\text{eq}} - T_{c0})|/T_{c0}$, the reduced value φ_{eq} of the equilibrium order parameter jump $|\psi|_{\text{eq}}$ and the latent heat Q of the equilibrium transition are given for films of different thicknesses and substances in table 2. The thicknesses are chosen to ensure the validity of the theory [3, 20] used in our analysis and to satisfy other important requirements presented in section 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 φ_{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 $L_0 = 0.05 \mu\text{m}$. The latent heat Q has maximal values for In, where the critical field is the highest for the considered materials.

4. Conclusion

Contrary to our initial expectations that films made of superconductors with extremely small GL parameter κ such as Al and, in particular, W will be the best candidates for an experimental search for the HLM effect, our careful analysis definitely gives a 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 to be inconvenient for the same reason because of their very low critical field H_{c0} . The importance of the critical magnetic field H_{c0} for the clearly manifested first-order phase transition has been established and discussed in section 3. Although In has ten times higher GL number κ 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 zero external magnetic field. We have also demonstrated for

the first time that the experiments can be most successfully performed in type I superconducting films with relatively high critical magnetic fields.

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