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## ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

# Effect of Surface Free Energy on Critical Field $H_{c3}$

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**Abstract**—The emergence of surface superconductivity in a type I superconductor is considered taking into account the surface free energy of the superconducting phase. It is shown that the disregard of the surface energy leads to a substantial error in determining the Ginzburg–Landau parameter from the measurements of the $H_{c3}$ field.

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It is generally assumed in the Ginzburg–Landau theory [1] that the surface free energy of the superconducting phase is negligible. For this reason, the following boundary condition is imposed on order parameter $\Psi$ in the description of surface superconductivity:

$$\Psi' = 0. \quad (1)$$

Here and below, the prime indicates the derivative with respect to the normal to the superconductor–vacuum interface. (We will henceforth use the London gauge in which the normal component of the vector potential at the interface is zero.)

The assumption concerning the smallness of the surface free energy was confirmed in [2], where the following boundary condition was derived:

$$\Psi' = \lambda_G \frac{\Psi}{\xi_0}, \quad (2)$$

where $\lambda_G \sim a_0/\xi_0 \ll 1$ has the order of the ratio of atomic spacing $a_0$ to the average size $\xi_0$ of a Cooper pair. For the superconductors considered here, this is a small quantity ($\lambda_G \approx 10^{-3}$).

Solution of the Ginzburg–Landau equations with boundary condition (1) leads to the well-known relation between critical field $H_{c3}$ of surface superconductivity and thermodynamic critical field $H_c$:

$$H_{c3}(\tau) = 1.695\kappa\sqrt{2}H_c(\tau), \quad (3)$$

where $\kappa$ is the Ginzburg–Landau parameter and $\tau = (T - T_c)/T_c$ is the reduced temperature. It should be emphasized that in the Ginzburg–Landau theory, the coefficient in this relation is independent of temperature. Therefore, ratio (3) of the critical fields can be used for experimental determination of parameter $\kappa$.

However, the results of measurements [3, 4] have shown that the $H_{c3}/H_c$ ratio for some superconductors (Al, In, Sn, and Pb) varies with temperature. This variation exceeds 10% even for $\tau \sim 10^{-2} \ll 1$, where no noticeable deviation from the Ginzburg–Landau theory should take place.

A natural way of eliminating this contradiction between the experimental results and the theory is to refine boundary condition (1). The attempts at using the BCS theory for this purpose (see the literature cited in [3]) have not led to a satisfactory result. The values of parameter $\kappa$ for the above superconductors have been determined with a low accuracy.

Here, we consider the variational boundary conditions determined by the surface free energy of the superconducting phase. We will write the surface free energy in the most general form corresponding to the Ginzburg–Landau theory.

The variational boundary conditions depend both on the surface free energy and the bulk free energy. Since we are interested in the role of the surface free energy, we will confine our analysis to a homogeneous superconductor whose bulk energy has the simplest form.

The bulk free energy density of a superconductor in a constant external field $H_0$ can be written in the form [5]

$$F_v = \alpha\tau|\Psi|^2 + \frac{\beta}{2}|\Psi|^4 + g\left|\left(\nabla - i\frac{2\pi}{\Phi_0}\mathbf{A}\right)\Psi\right|^2 + \frac{B^2}{8\pi} - \frac{BH_0}{4\pi}. \quad (4)$$

Here, $\Phi_0$ denotes the flux quantum, $B$ is the magnetic field induction in the superconductor, and $\mathbf{A}$ is the vector potential. Let us now pass to dimensionless variables. For this purpose, we will use the notation of the Ginzburg–Landau theory,

$$\xi_0 = \sqrt{\frac{g}{\alpha}}, \quad \kappa = \frac{\Phi_0}{2\pi}\sqrt{\frac{\beta}{8\pi g^2}}, \quad (5)$$

and natural units of length $\xi(\tau)$, magnetic field $H_{c2}(\tau)$, and equilibrium value of the order parameter $\Psi_0(\tau)$:

$$\xi(\tau) = \frac{\xi_0}{\sqrt{-\tau}}, \quad H_{c2}(\tau) = \frac{\Phi_0}{2\pi\xi_0^2}(-\tau), \quad |\Psi_0(\tau)|^2 = \frac{\alpha}{\beta}(-\tau). \tag{6}$$

In the dimensionless variables

$$\psi = \frac{\Psi}{|\Psi_0|}, \quad \mathbf{a} = \frac{\mathbf{A}}{H_{c2}\xi}, \quad b = \frac{B}{H_{c2}}, \quad h_0 = \frac{H_0}{H_{c2}}, \tag{7}$$

bulk energy density (4) has the form

$$F_v = \frac{\alpha^2}{\beta}\tau^2\left\{-|\psi|^2 + \frac{1}{2}|\psi|^4 + |\nabla\psi - i\mathbf{a}\psi|^2 + \kappa^2(b^2 - 2bh_0)\right\}. \tag{8}$$

It should be noted that $F_v$ is the sum of the second-order invariants in $\tau$.

Omitting dimensional common factor $\alpha^2\tau^2/\beta$, we can calculate the free energy of the superconducting phase ($|\psi| = 1$, $b = 0$) equal to $-1/2$ and the free energy of the normal phase ($|\psi| = 0$, $b = h_0$) amounting to $-\kappa^2 h_0^2$. The condition for coexistence of the phases is satisfied in the thermodynamic critical field $h_0 = h_c = 1/\kappa\sqrt{2}$. Since quantity $h_0$ in the problem under investigation is a constant, we can measure the free energy of the superconductor from the energy of the normal phase; for this purpose, we add quantity $\kappa^2 h_0^2$ to the last term in expression (8):

$$\mathcal{F}_v = -|\psi|^2 + \frac{1}{2}|\psi|^4 + |\nabla\psi - i\mathbf{a}\psi|^2 + \kappa^2(b - h_0)^2. \tag{9}$$

We can write the surface free energy in the form of an expansion in $\Psi_s$, viz., the value of order parameter $\Psi$ at the boundary of the superconductor:

$$F_s = (C_0 + C_1\tau)|\Psi_s|^2 + \frac{D_1}{2}|\Psi_s|^4 + G_1\left|\left(\frac{\partial}{\partial l} - i\frac{2\pi}{\Phi_0}A_l\right)\Psi_s\right|^2. \tag{10}$$

Here, index $l$ indicates the components of the gradient and vector potential directed in the plane of the interface. This expression contains the first-order term in $\tau - C_0|\Psi_s|^2$. It was shown in [2], however, that coefficient $C_0$ is small. Therefore, retaining the second-order terms in $\tau$ is not an overestimate.

We assume that the external field direction lies in the plane of the interface. Using the standard gauge procedure for the 1D problem (along $x$), in which vector potential $\mathbf{a} = a(x)\mathbf{e}_y$ also lies in this plane and order parameter $\psi(x) = f(x)$ is real-valued, we can write the total energy in the form

$$E = \int_0^\infty dx\left[(f')^2 + a^2f^2 - f^2 + \frac{1}{2}f^4 + \kappa^2(b - h_0)^2\right] + f^2(0)\left\{\frac{\lambda_0}{\sqrt{-\tau}} + [-\lambda_1 + \gamma_1 a^2(0) + \frac{\eta_1}{2}f^2(0)]\sqrt{-\tau}\right\}, \tag{11}$$

where

$$\lambda_0 = \frac{C_0}{\alpha\xi_0}, \quad \lambda_1 = \frac{C_1}{\alpha\xi_0}, \quad \gamma_1 = \frac{G_1}{g\xi_0}, \quad \eta_1 = \frac{D_1}{\beta\xi_0}. \tag{12}$$

The temperature dependence of the surface free energy in expression (11) is a consequence of reduction to the dimensionless form, in which the Ginzburg–Landau energy (the integrand) is independent of temperature.

The minimum of functional (11) corresponds to the Ginzburg–Landau equations

$$f'' = f(a^2 - 1 + f^2), \quad a' = b, \quad b' = \frac{f^2}{\kappa^2}a \tag{13}$$

with boundary conditions

$$f'(0) = \lambda f(0) + \eta_1\sqrt{-\tau}f^3(0), \quad b(0) = h_0 + \gamma_1\sqrt{-\tau}\frac{f^2(0)}{\kappa^2}a(0), \tag{14}$$

where

$$\lambda = \frac{\lambda_0}{\sqrt{-\tau}} - \lambda_1\sqrt{-\tau} + \gamma_1\sqrt{-\tau}a^2(0). \tag{15}$$

We have obtained the boundary conditions in the general form, which depend on four parameters (12). One of these parameters (the De Gennes parameter $\lambda_0$) is especially small. We use the notation $\lambda_0$ instead of $\lambda_G$ from condition (2) to emphasize that the sign and magnitude of parameter $\lambda_0$ may noticeably differ from the value of $\lambda_G$, which is an order-or-magnitude estimate. A reasonable expression for $\lambda_0$ was derived in [6]:

$$\lambda_0 \approx \frac{a_0}{\xi_0\sqrt{2}}\frac{1}{\Lambda^2} \times \int_0^\infty dx\frac{N(x)}{N(\infty)}[V(x)N(x) - V(\infty)N(\infty)], \tag{16}$$

where $N(x)$ is the local density of states at the Fermi level, $V(x)$ is the Cooper interaction constant, and $\Lambda = V(\infty)N(\infty)$ ($\Lambda \approx 0.2$–$0.3$).

Parameter $\gamma_1$ defines the surface current and the field jump at the boundary of the superconductor,

which can be explained by the nonlocal nature of the relation between the superconducting current and the vector potential. For this reason, we refer to $\gamma_1$ as the Pippard parameter. It should be emphasized that quantitative analysis of parameters (12) is beyond the scope of the phenomenological model considered here and can be carried out only in terms of microscopic theory.

Critical field $h_{sc}$ in which surface superconductivity appears can be determined from the linearized eigenvalue problem

$$f'' = f(a^2 - 1) \tag{17}$$

with the boundary condition

$$f'(0) = \lambda f(0), \tag{18}$$

where vector potential $a(x) = h_0(x - x^*)$ corresponds to the unperturbed field. Parameter $x^*$ has the meaning of the coordinate at which the superconducting current changes its sign and is chosen so that field $h_{sc}$ is maximal.

A distinguishing feature of problem (17), (18) is that parameter $\lambda$ cannot be treated as an independent quantity. In accordance with relation (15), this parameter depends on the value of $a(0)$, which in turn is determined from the solution to Eq. (17). Therefore, the solution to problem (17), (18) shown in Fig. 1 is a function of two variables: $h_{sc} = \mathcal{H}(\lambda, \varepsilon)$, where $\varepsilon = \gamma_1\sqrt{-\tau}$.

For $\varepsilon = 0$, dependence $\mathcal{H}(\lambda, 0)$ was obtained in [7]. It is depicted by curve *1* in Fig. 1. The value of $\lambda = 0$ corresponds to the field $h_{sc} = h_{c3} = 1.695$; for $|\lambda| \ll 1$, the approximation $h_{sc} \approx h_{c3}(1 - \lambda)$ holds. It should also be noted that for $\lambda > 0$, field $h_{sc} < h_{c3}$; i.e., surface superconductivity is suppressed, and when $\lambda \longrightarrow +\infty$, the field tends to $h_{sc} \longrightarrow h_{c2} = 1$.

To derive the temperature dependence $h_{sc}(\tau)$, we must determine dependence $\lambda(\tau)$. For this, we must find the value of $a(0)$ in formula (15). This can be done using the value of the first integral of the Ginzburg–Landau equations,

$$(f')^2 + \kappa^2(b^2 - h_0^2) = a^2f^2 - f^2 + \frac{1}{2}f^4, \tag{19}$$

for $x = 0$. Substituting boundary conditions (14) and passing to the limit $f(0) \longrightarrow 0$, we obtain

$$a(0) = h_0\varepsilon - \sqrt{1 + \lambda^2 + h_0^2\varepsilon^2}. \tag{20}$$

Eliminating $a(0)$ from expression (15), we obtain the fourth-degree equation in $\lambda$:

$$\left[\lambda^2 - \frac{\lambda}{\varepsilon} + K\right]^2 = 4h_0^2\varepsilon^2(1 + \lambda^2 + h_0^2\varepsilon^2),$$
$$K = \frac{\lambda_0}{\gamma_1(-\tau)} + 1 - \frac{\lambda_1}{\gamma_1} + 2h_0^2\varepsilon^2, \tag{21}$$

which defines the $\lambda(\tau)$ dependence implicitly.

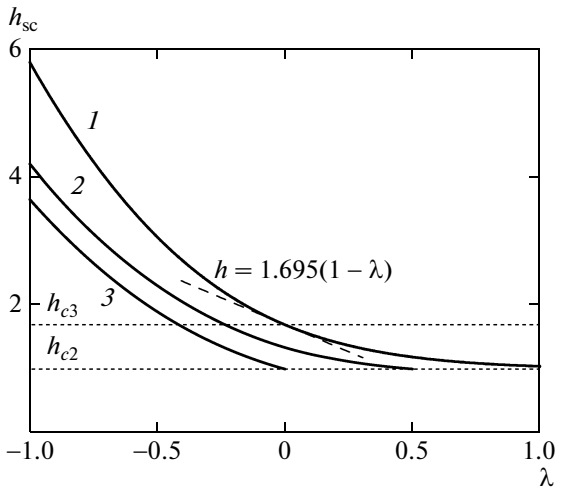


**Fig. 1.** Dependence of field $h_{sc} = \mathcal{H}(\lambda, \varepsilon)$ on parameter $\lambda$ for several values of $\varepsilon$: (*1*) $\varepsilon = 0$, (*2*) $\varepsilon = 1$, and (*3*) $\varepsilon \longrightarrow \infty$.

For $h_0\varepsilon \ll 1$, we can use the approximate relation $a(0) \approx -\sqrt{1 + \lambda^2}$ and lower the equation to the second degree. The solution to this equation,

$$\lambda(\tau) = \frac{1 - \sqrt{1 - 4\gamma_1\lambda_0 - \tau 4\gamma_1(\lambda_1 - \gamma_1)}}{2\gamma_1\sqrt{-\tau}}, \tag{22}$$

gives the required temperature dependence $h_{sc}(\tau) = \mathcal{H}(\lambda(\tau), 0)$.

In expression (22), there is a characteristic temperature,

$$\tau^* = \frac{1}{4\gamma_1(\lambda_1 - \gamma_1)}, \tag{23}$$

which separates two ultimate cases. For $(-\tau) \ll \tau^*$ (and $4\gamma_1|\lambda_0| \ll 1$), expression (22) can be simplified,

$$\lambda(\tau) \approx \frac{1}{\sqrt{-\tau}}\lambda_0 - \sqrt{-\tau}(\lambda_1 - \gamma_1), \tag{24}$$

which corresponds to the value of $a(0) = -1$. The other case $(-\tau) \gg \tau^*$,

$$\lambda(\tau) \approx -\sqrt{\frac{\lambda_1 - \gamma_1}{\gamma_1}}, \tag{25}$$

leads to a linear dependence $H_{sc}(\tau)$, which has a larger slope than that of $H_{c3}(\tau)$.

For $h_0\varepsilon \gg \sqrt{1 + \lambda^2}$, relation (20) leads to

$$a(0) \approx -\frac{1 + \lambda^2}{2h_0\varepsilon} \longrightarrow 0, \tag{26}$$

which allows us to omit the last term in formula (15). In this case, we obtain temperature dependence $h_{sc}(\tau) = \mathcal{H}(\lambda(\tau), \infty)$, where

$$\lambda(\tau) \approx \frac{\lambda_0}{\sqrt{-\tau}} - \lambda_1\sqrt{-\tau}. \tag{27}$$

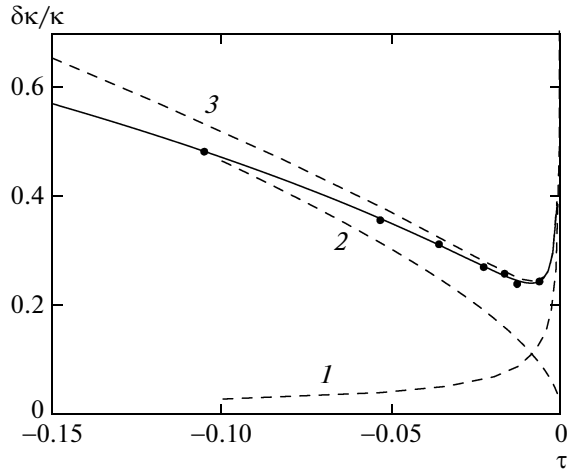


**Fig. 2.** Dependence of the relative variation of $\kappa_{sc}$ on reduced temperature $\tau$ for different values of $\lambda(\tau)$. The solid curve is calculated by formula (22) with parameters $\lambda_0 = -9.1 \times 10^{-3}$, $\lambda_1 = 1.76$, and $\gamma_1 = 0.72$. The dashed curves correspond to the following approximations: (*1*) $\lambda_0/\sqrt{-\tau}$, (*2*) $\sqrt{-\tau}\,(\gamma_1 - \lambda_1)$; (*3*) formula (24). Circles correspond to the results obtained in [3] for tin.

The above arguments correctly describe the $\lambda(\tau)$ dependence; however, numerical approximation should be used for function $\mathcal{H}(\lambda, \varepsilon)$. In addition, for $h_0\varepsilon \sim 1$, the $h_{sc}(\tau)$ dependence can be obtained only by solving problem (17), (18) numerically.

To compare our results with experiment, it is convenient to introduce (as was done in experimental works) the effective value of $\kappa_{sc}(\tau)$ and its deviation from parameter $\kappa$:

$$\kappa_{sc}(\tau) = \frac{h_{sc}(\tau)}{h_{c3}}\kappa, \quad \frac{\delta\kappa(\tau)}{\kappa} = \frac{h_{sc}(\tau) - h_{c3}}{h_{c3}}. \qquad (28)$$

These expressions are convenient because the following relation holds for small $\lambda$ and $\varepsilon$:

$$\frac{\delta\kappa(\tau)}{\kappa} \approx -\lambda(\tau). \qquad (29)$$

Figure 2 shows dependence $\delta\kappa(\tau)/\kappa$ for tin based on the results from [3], which was obtained using model (22) with parameters $\lambda_0 = -9.1 \times 10^{-3}$, $\lambda_1 = 1.76$, and $\gamma_1 = 0.72$. It is important that in view of the slow decrease of function $1/\sqrt{-\tau}$, the presence of the second term in formula (24), which is proportional to $\sqrt{-\tau}$, is not clearly seen in the figure. Conversely, it can be seen from the figure that the left part of the curve is a weakly curved linear dependence, which can be "smoothly" extrapolated to $\tau = 0$. It follows from the figure, however, that such an extrapolation gives an estimate for $\kappa$ higher by 20%. Indeed, from the extrapolation to $\tau = 0$, the value of $\kappa \approx 0.093$ was obtained in [3] for tin, while approximation (22) gives $\kappa \approx 0.075$. The $\kappa_{sc}(\tau)$ dependence for tin obtained in [4] noticeably differs from that reported in [3]; nevertheless, we obtained a close value $\kappa \approx 0.073$.

Since the value of $h_0\varepsilon$ in our approximations varies in the range $0.1 < h_0\varepsilon < 0.7$ and the condition $h_0\varepsilon \ll 1$ for which expression (22) was obtained is violated, we have also determined parameter $\kappa$ using the direct solution to problem (17), (18). As a result, we obtained a slightly higher value of $\kappa \approx 0.079$.

Too high values of parameter $\kappa$ were also obtained for indium by extrapolating $\kappa_{sc}(\tau)$ to $\tau = 0$. The extrapolation in [3] gives $\kappa \approx 0.062$, while approximation (22) gives $\kappa \approx 0.05$.

It should be noted that the experimental works were aimed at analysis of the $\kappa_{sc}(\tau)$ dependence for its subsequent extrapolation to $\tau = 0$. But the behavior of the curve in close vicinity to $T_c$ was ascribed to size effects, so the detailed measurements in this temperature range were not performed.

Recent studies devoted to surface superconductivity of lead [8], in which measurements were taken in the immediate vicinity of $T_c$, have revealed good agreement between the observed $h_{sc}(\tau)$ dependence and the model described here. The value of the Ginzburg–Landau parameter obtained in [8] for lead is $\kappa \approx 0.2$.

Boundary condition (18), (15) does not contain parameter $\eta_1$. This is not surprising because in the linear problem, we disregard the term proportional to $f^4(0)$. However, this parameter can also be determined experimentally. As a matter of fact, the following two scenarios are possible in field $h_{sc}(\tau)$: (i) a localized surface state is formed, which is stable in a certain range of fields and temperatures, or (ii) an unstable state is formed, and superconductivity immediately sets in the entire sample. If we disregard the surface free energy, the choice of the scenario is determined by the value of parameter $\kappa$. This problem was considered in [9], in which the critical value of Ginzburg–Landau parameter $\kappa_F \approx 0.405$ (Feder point) was determined. For $\kappa < \kappa_F$, bulk superconductivity appears in field $h_{c3}$, while for $\kappa > \kappa_F$, stable surface states exist [10].

If the surface free energy differs from zero, the Feder critical point depends not on the Ginzburg–Landau parameter only, but also on parameters (12) of the surface free energy. Let us consider the stability of the surface state near the $h_{sc}(\tau)$ curve for $f \longrightarrow 0$. This can be done in the simplest way by evaluating the energy $E_0$ at the extremum of functional (11). For this purpose, we integrate term $(f')^2$ in relation (11) by parts and substitute expression (13) for $f''(x)$ and (14) for $f'(0)$:

$$E_0 = \int_0^\infty \left[\kappa^2(b - h_0)^2 - \frac{1}{2}f^4\right]dx - \frac{\eta_1}{2}f^4(0). \qquad (30)$$

Since $f \longrightarrow 0$ and $(b - h_0) \propto f^2$ as the $h_{sc}(\tau)$ curve is approached, energy (30) vanishes as $E_0 \approx cf^4(0)$. The sign of coefficient $c$ changes at the critical point. Therefore, we can express $\eta_1$ in terms of solution $a(x)$, $f(x)$ to linear problem (17), (18):

$$\eta_1 = \frac{1}{f^4(0)}\int_0^\infty \left[\frac{2}{\kappa^2}u^2(x) - f^4(x)\right]dx,$$
$$u(x) = \int_x^\infty a(\tilde{x})f^2(\tilde{x})d\tilde{x}. \quad (31)$$

Thus, knowing the position of the Feder point on the $h_{sc}(\tau)$ curve and the three parameters determining boundary condition (15) for the linear problem, we can also determine the value of parameter $\eta_1$. By way of illustration of the effect of parameters (12) on the critical value $\kappa_F$, Fig. 3 shows the $\kappa_F(\lambda)$ dependence for $\gamma_1 = \eta_1 = 0$.

If we consider the surface free energy as a temperature correction to the Ginzburg–Landau energy, the question of the next-order corrections in $\tau$ arises. These are the corrections to volume energy density (4) on the order of $\tau^3$. These corrections were considered in [11], in which it was shown that for $(-\tau) \ll 1$, such corrections lead to the dependence

$$\frac{\delta\kappa(\tau)}{\kappa} \approx 0.41(-\tau). \quad (32)$$

As expected, the value of the effects considered here is much larger than the value given by formula (32) in the vicinity of $T_c$.

Since the thickness of the surface state is on the order of $\xi(\tau)$ and is proportional to $1/\sqrt{-\tau}$, we can conclude that the corrections to the volume energy lead to integral powers of $\tau$ in the $\kappa_{sc}(\tau)$ dependence, while the terms in the surface energy lead to half-integral powers of $\tau$. This conclusion is in conformity with the results obtained in the BCS theory. In particular, the square root dependence $\kappa_{sc}(\tau)$ was obtained in [12] from analysis of the conditions of electron reflection from the interface with vacuum.

The last question to be considered here concerns the surface superconducting transition temperature in zero field. At temperatures higher than $T_c$, the volume energy changes its sign and becomes positive, while the surface free energy remains negative (for $\lambda_0 < 0$) and finite. Therefore, the total energy of the surface state remains negative up to a certain temperature $\tau_s$, at which superconductivity disappears [13]. It can be shown that $\tau_s$ satisfies the equation

$$\tau_s = (\lambda_0 + \lambda_1\tau_s)^2 \lesssim \lambda_0^2. \quad (33)$$

The experimental observation of surface superconductivity above $T_c$ appears to be difficult in view of the small value of parameter $\lambda_0$.

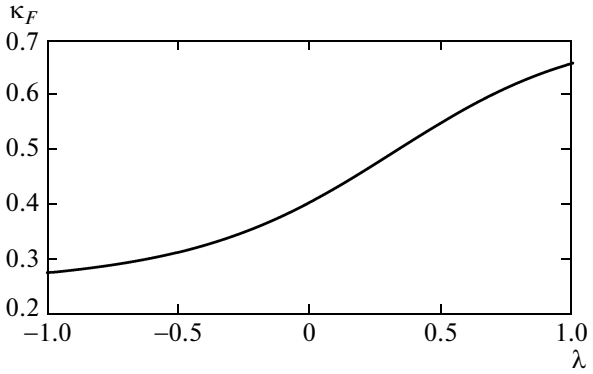


**Fig. 3.** Dependence of the critical value of $\kappa_F(\lambda)$ for $\gamma_1 = \eta_1 = 0$. The value of $\lambda = 0$ corresponds to $\kappa_F \approx 0.405$.

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