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Critical dynamics of inhomogeneous superconducting films*

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The Gaussian-random model for the critical dynamics in inhomogeneous superconducting films is augmented by including the quartic interaction term in the free-energy functional. Within the Hartree approximation a self-consistent calculation of the order-parameter relaxation rate Γ is presented. The temperature dependence of Γ exhibits a pronounced change for various values of the Ginzburg critical width. A comparison with an experiment on Al-Al_2O_3-Pb junction is presented and the validity of the Gaussian-random model for this specific experiment is demonstrated.

I. INTRODUCTION

In recent years inhomogeneous superconducting films are becoming the subject of increasing interest from both theoretical[1-3] and experimental[4-6] points of view.

A suitable starting point for studying the static properties of an inhomogeneous superconducting film is a generalization of the usual Ginzburg-Landau free-energy functional which takes into account local fluctuations of the BCS coupling strength due to the structural inhomogeneities of the film:

$$F(\psi) = d \int dx^2 [A(\vec{x})|\psi|^2 + \tfrac{1}{2}B|\psi|^4 + C|\vec{\nabla}\psi|^2], \quad (1.1)$$

where $\psi(\vec{x})$ is the order parameter, d is the thickness of the film, $A(x) = A_0 + \delta A(\vec{x})$, $\delta A(\vec{x})$ being proportional to the local fluctuations of T_c, $A_0 = N_0(T/T_{c0} - 1) = N_0\epsilon$, $B = 0.106 N_0/T_{c0}^2$, $C = N_0\xi(0)^2$, where $\xi(0)$ is the temperature-independent correlation length in a homogeneous film, and N_0 is the bulk density of states at the Fermi level.

Of particular interest, connected with the dynamics of the order parameter, is the experiment of Anderson *et al.*,[4] in which a nonlinear temperature dependence of the relaxation rate Γ was found in contradiction with what one expects from a theory of classical slowing down.[7] The first explanation of such nonlinear temperature dependence was given in Ref. 3, using a generalization of Eq. (1.1) to a time-dependent order parameter in the Gaussian approximation [quartic term in Eq. (1.1) neglected].

The purpose of the present paper is to investigate the role of the quartic term on the critical dynamics within the Hartree approximation with the emphasis on detailed comparison with the above-mentioned experiment. Moreover, our calculations enable us to predict the degree of the nonlinearity of Γ as a function of the Ginzburg critical width of the film. Such nonlinearities could be verified by experiments on films with proper mean-free path l and film thickness d.

II. HARTREE-RANDOM MODEL

Generalizing the phenomenological theory of Shapero *et al.*[1] to time-dependent susceptibility $\chi(\vec{x}, \vec{x}', t)$, we have, in the Hartree approximation

$$\left(\frac{\partial}{\partial t} t + \gamma_0[A(\vec{x}) + B\langle\psi^*(\vec{x}, t)\psi(\vec{x}, t)\rangle + C\vec{\nabla}^2]\right)\chi(\vec{x}, \vec{x}', t) = \frac{1}{d} T\gamma_0 \delta(\vec{x} - \vec{x}')\delta(t), \quad (2.1)$$

where $\gamma_0 = 8T_{c0}/N_0\pi$.

Solving Eq. (2.1) for $\chi(\vec{x}, \vec{x}', t)$, treating $\delta A(\vec{x})$ as a small perturbation, and configurationally averaging $\chi(\vec{x}, \vec{x}', t)$, we define an averaged translational invariant $G(\vec{x} - \vec{x}', t)$ as

$$G(\vec{x} - \vec{x}', t) = \langle\chi(\vec{x}, \vec{x}', t)\rangle_c, \quad (2.2)$$

which obeys the diagrammatic equation in Fig. 1. In Fig. 1, the full thin lines represent the dynamic susceptibility of a homogeneous superconductor, which in the Gaussian model ($B = 0$; $A = A_0$) is given by

$$G_0^{-1}(\vec{q}, \omega) = \frac{Cd}{T}\left(\vec{q}^2 + \frac{A_0}{C} - \frac{i\omega}{\gamma_0 C}\right). \quad (2.3)$$

The full dynamic susceptibility [Fourier transform of Eq. (2.2)] is given by

$$G^{-1}(\vec{q}, \omega) = G_0^{-1}(\vec{q}, \omega) + \Sigma(\vec{q}, \omega), \quad (2.4)$$

where the self-energy $\Sigma(\vec{q}, \omega)$ is, according to Fig. 1,

$$\Sigma(\vec{q}, \omega) = \frac{d}{T} B G(\vec{x} = \vec{x}', t = 0) - \frac{d^2}{T^2}\int \frac{d^2 q'}{(2\pi)^2} \tilde{S}(\vec{q} - \vec{q}') G(\vec{q}', \omega). \quad (2.5)$$

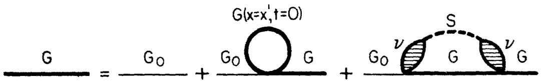

FIG. 1. Diagrammatic Dyson-type equation for the configurational-averaged dynamic susceptibility G, in Hartree-random model.

In Eq. (2.5), $\tilde{S}(\vec{q}) = S(q)\nu(q)^2$, where $S(q)$ is the Fourier transform of the correlation function of $\delta A(\vec{x})$,

$$\langle \delta A(\vec{x})\delta A(\vec{x}')\rangle_c = N_0^2(\langle \delta T_c^2\rangle / T_{c0}^2)\exp(-\alpha^2|\vec{x}-\vec{x}'|^2), \tag{2.6}$$

and $\nu(q)$ represents the screening of $\delta A(\vec{x})$ fluctuations due to the quartic term. In two dimensions, the expression for $\nu(q)$ is given by[8]

$$\nu(q)^{-1} = 1 + \frac{T}{T_{c0}}\frac{w}{\kappa^2\xi(0)^2}\frac{\ln\{q/2\kappa + [1+(q/2\kappa)^2]^{1/2}\}}{(q/2\kappa)[1+(q/2\kappa)^2]^{1/2}}. \tag{2.7}$$

In Eq. (2.7), $w = 0.106/4\pi N_0 d[\xi(0)]^2 T_{c0}$ is related to the Ginzburg critical width,[9] and $\kappa(T)$ is the temperature-dependent inverse correlation length defined by

$$[\kappa(T)]^2 = [\xi(0)]^{-2}\left(\epsilon + \frac{T}{N_0 d}\Sigma(0,0)\right). \tag{2.8}$$

Approximating $\Sigma(\vec{q}', \omega)$ by $\Sigma(q=0,\omega)$ in the expression of $G(\vec{q}', \omega)$ inside the integral of Eq. (2.5), we obtain the following self-consistent equation for the zero-momentum self-energy:

$$\Sigma(0,\omega) = (d/T)BG(\vec{x}=\vec{x}', t=0) - \frac{d^2}{T^2}N_0^2\frac{\langle\delta T_c^2\rangle}{T_{c0}^2}\int_0^\infty \frac{(q/2\alpha^2)dq\,\exp(-q^2/4\alpha^2)\nu(q)^2}{(Cd/T)(q^2+\kappa^2 - i\omega/\gamma_0 C) + \hat{\Sigma}(0,\omega)}, \tag{2.9}$$

where

$$\hat{\Sigma}(0,\omega) = \Sigma(0,\omega) - \Sigma(0,0). \tag{2.10}$$

Using Eqs. (2.4) and (2.8), we calculate the quantity $G(\vec{x}=\vec{x}', t=0)$ as follows:

$$G(\vec{x}=\vec{x}', t=0) = \int_0^{q_D}\frac{q\,dq}{2\pi}G(q,0) = (T/4\pi Cd)\ln(1+q_D^2/\kappa^2), \tag{2.11}$$

where q_D is a Debye-like cutoff π/a, with a equal to the forces range $\xi(0)$. Letting $\omega = 0$ in Eq. (2.9), and again using Eq. (2.8), we obtain the self-consistent equation for the dimensionless quantity $X = \kappa(T)\xi(0)$,[10]

$$X^2 = \epsilon + T/T_{c0}w\ln\{1+q_D^2[\xi(0)]^2/X^2\} - (r_2/r_1)[e^Y E_1(Y) - I_s(Y)], \tag{2.12}$$

where the parameters r_1 and r_2 are defined as follows:

$$r_1 = 4\alpha^2[\xi(0)]^2$$

and

$$r_2 = \langle\delta T_c^2\rangle / T_{c0}^2. \tag{2.13}$$

The expression for the quantity $I_s(Y)$ is given by the integral

$$I_s(Y) = \int_0^\infty \frac{e^{-z}dz}{z+Y}\left(1 - \frac{1}{[1+\Pi_0(z,Y)]^2}\right), \tag{2.14}$$

where $Y = X^2/r_1$ and[8]

$$\Pi_0(z,Y) = \frac{T}{T_{c0}}\frac{w}{Yr_1}\frac{1}{u(1+u^2)^{1/2}}\ln[u+(1+u^2)^{1/2}]; \quad u = (z/4Y)^{1/2}. \tag{2.15}$$

Equations (2.12)–(2.15) define the self-consistent Hartree-random(HR) model.

The Gaussian-random(GR) model approximation follows from Eqs. (2.12), (2.14), and (2.15), by putting $w = 0$. The resulting self-consistent equation for X in GR model takes the form

$$X_G^2 = \epsilon - (r_2/r_1)e^{Y_G}E_1(Y_G); \quad Y_G = X_G^2/r_1. \tag{2.16}$$

During the numerical process of solving the self-consistent equation (2.12) for X, we perform a numerical calculation of the integral $I_s(Y)$, which represents the screening correction due to the quartic term.

III. ORDER-PARAMETER RELAXATION RATE

The HR-model expression for the relaxation time τ is calculated using the formula[11]

$$\tau = \tau_{q=0} = \lim_{\omega\to 0}\left(\frac{G(0,\omega) - G(0,0)}{i\omega G(0,0)}\right). \tag{3.1}$$

This expression, after some rearrangements, takes the form

$$\tau = \left(\frac{d}{\gamma_0 T}G(0,0)\right)\left(1 - \frac{T\gamma_0}{d}\frac{\partial\Sigma(0,\omega)}{\partial(i\omega)}\bigg|_{\omega=0}\right). \tag{3.2}$$

The first factor on the right-hand side of the Eq. (3.2) represents the conventional slowing down τ_{conv}, and is proportional via $G(0,0)$ [see Eq. (2.4)] to $X^{-2} = [\kappa(T)\xi(0)]^{-2}$. With the remark that $\partial\hat{\Sigma}(0,\omega)/$

$\partial\omega = \partial\Sigma(0,\omega)/\partial\omega$, we perform the derivative of both sides of the self-consistent equation (2.9) for $\Sigma(0,\omega)$. Taking the limit $\omega \to 0$, we obtain, after a slight rearrangement, the following expression for the relaxation rate $\Gamma = \tau^{-1}$:

$$\frac{\Gamma}{T_{c0}} = \frac{8}{\pi} X^2 \left[1 - \frac{r_2}{r_1^2}\left(\frac{1}{Y} - e^Y E_1(Y) - I_D(Y)\right)\right], \qquad (3.3)$$

where the expression for $I_D(Y)$ is given by

$$I_D(Y) = \int_0^\infty \frac{e^{-z} dz}{(z+Y)^2}\left(1 - \frac{1}{[1+\Pi_0(z,Y)]^2}\right). \qquad (3.4)$$

Solving Eq. (2.12) numerically for X, and performing once again a numerical calculation of the integral $I_D(Y)$, we obtain the full relaxation rate which describes the critical slowing down of the order parameter.

Before going to the numerical results, we would like to exhibit the different limits of the expression (3.3).

(i) Putting $r_2 = 0$ and $w = 0$ in Eq. (2.13), we obtain the Gaussian result for a homogeneous superconductor

$$\frac{\Gamma_0}{T_{c0}} = \frac{8}{\pi}\left(\frac{T}{T_{c0}} - 1\right). \qquad (3.5)$$

(ii) Using the self-consistent equation for X_G in GR model [Eq. (2.16)], we derive the following analytical expression for the GR-model relaxation rate:

$$\frac{\Gamma_G}{T_{c0}} = \frac{8}{\pi} X_G^2 \left[1 - \frac{1}{r_1}\left(\frac{r_2}{X_G^2} + X_G^2 - \epsilon\right)\right], \qquad (3.6)$$

where the factor in front of the large square brackets represents the conventional slowing down of the GR model.[3] Since we intend to use this model as a starting one for the HR model, we briefly discuss its important features. Being interested in the very vicinity of $T_c (\epsilon \lesssim 5\times10^{-2})$, where $X^2 \ll 1$, we approximate in Eq. (2.16) $e^Y E_1(Y)$ by $\ln(1/Y) - \gamma + O(Y\ln(1/Y))$, where γ is Euler's constant.[10] We define a new variable $Z_G = X_G^2(r_1/r_2)$, which obeys the equation

$$Z_G = \ln(Z_G) + \left[\frac{r_1}{r_2}\epsilon + \ln\left(\frac{r_2}{r_1^2}\right) + \gamma\right]. \qquad (3.7)$$

The condition for the existence of a positive solution Z_G of Eq. (3.7) *above* some temperature T_c is that at this temperature $(T = T_c)$, the term inside the large square brackets of the right-hand side of Eq. (3.7) will be equal to 1; i.e., at $T = T_c$, $(Z_G)_{\min} = 1$, and the shift of the bare transition temperature T_{c0} is given by

$$\epsilon_c = \frac{T_c}{T_{c0}} - 1 = \frac{r_2}{r_1}\left[1 - \ln\left(\frac{r_2}{r_1^2}\right) - \gamma\right]. \qquad (3.8)$$

A finite solution at $T = T_c$ for Z_G $(=1)$ determines a finite conventional slowing-down rate $(8/\pi) r_2/r_1$, and within the logarithmic approximation it is easy to show that Γ_G approaches zero at this temperature. Moreover, taking the derivative of the Eq. (3.7) with respect to T, and using the fact that at $T = T_c$, $Z_G = 1$, we can see that the slope of X_G^2 $(\sim Z_G)$ is infinite at this temperature[3] with a critical exponent $\frac{1}{2}$.[12] The numerical results described in the following chapter confirm all of those features.

IV. NUMERICAL RESULTS AND COMPARISON WITH THE EXPERIMENT

Choosing reasonable values for the parameters r_1 and r_2, we calculate first the GR-model result for Γ_G/T_{c0}. Using $\alpha\xi(0) = 0.943$ $(r_1 = 3.63)$[13] and $r_2 = \langle\delta T_c^2\rangle/T_{c0}^2 = 2.25\times10^{-2}$ the expression (3.8) gives the shift of T_{c0} due to the inhomogeneity of the film. The value $\epsilon_c = 4.3\times10^{-2}$ obtained using the formula (3.8) agrees very well with the exact numerical calculation of the relaxation rate in GR model exhibited by curve (a) in Fig. 2. The slope of $\Gamma_G(\epsilon)$ at $\epsilon = \epsilon_c$ is indeed infinite.

In order to see the influence of the quartic term (with the screening) we have to chose relative small values for $\xi(0)$ and d, in order to obtain a large Ginzburg critical width . From the explicit form of the expression for $\Pi_0(z,Y)$ [Eq. (2.15)], we can see that at $u = 0$, $\Pi_0(0,Y)$ reaches the maximum value which is equal to w/X^2. Larger values of w lead to larger values of Π_0 and consequently to larger values of the integral $I_s(Y)$. Then, through Eq. (2.12), this will result in a smaller value of X for a given ϵ. Hence, there is a positive-feedback mechanism which is responsible for a shift of $\Gamma(\epsilon)$

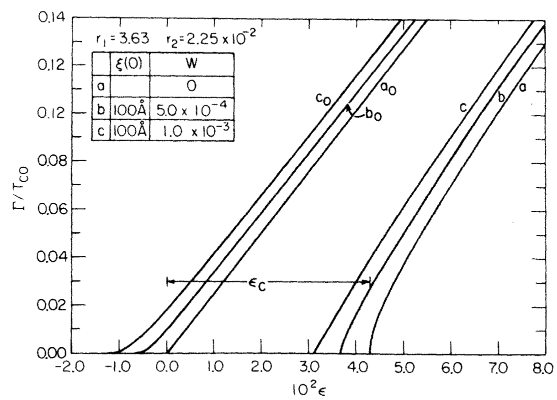

FIG. 2. Order parameter relaxation rate Γ, as a function of the reduced temperature $\epsilon = T/T_{c0} - 1$. The curve a (a_0) represents the Gaussian result for an inhomogeneous (homogeneous) film. The curves b, c (b_0, c_0) are the Hartree results for an inhomogeneous (homogeneous) film. The parameters r_1, r_2 and w, and $\xi(0)$ are defined in the text.

curve towards lower temperatures, thus cancelling partly the shift in the opposite direction due to the inhomogeneity. Moreover, the slope at the actual T_c is decreasing, and the characteristic knee of the GR model progressively disappears. This could be understood intuitively by looking at the homogeneous Hartree results (curves b_0 and c_0 in Fig. 2, which show the well-known leveling off characteristic to the Hartree approximation without randomness.

The importance of the quartic term decreases with decreasing values of w. Films with large degree of disorder (small values of the mean-free path l) will have larger values of w and will allow for a large effect of the quartic term. At the same time, the ratio between the correlation length of the frozen disorder (α^{-1}) and $\xi(0) \sim l^{1/2}$ will increase, and by inspection of the Eqs. (2.11) and (3.6) we can see that the GR-model features will tend to persist; i.e., bigger shift of T_{c0} accompanied by a stronger bending of the $\Gamma(\epsilon)$ curve. In the discussion above, we have assumed a fixed value of $\langle \delta T_c^2 \rangle / T_{c0}^2$. However, from the experimental point of view it should be mentioned that the parameters l, $\langle \delta T_c^2 \rangle$, and the ratio $\alpha^{-1}/\xi(0)$ are not independent, since they are all related to the structure of the disordered films.

Of particular interest is the experiment of Anderson *et al.*[4] on $Al - Al_2O_3 - Pb$ junctions, where the relaxation rate of the superconducting order parameter was obtained by measuring the excess fluctuation conductivity using the $I-V$ curves. We would like to compare our results with the data of the junction Al – 6 of Ref. 4 which is characterized by $l \simeq 20$ Å (which gives $\xi(0) \simeq 500$ Å), and thickness $d = 1300$ Å. With the latter values and $N_0 = 2\times 10^{18}$ $\text{cm}^{-3}\,{}^\circ\text{K}^{-1}$, and taking $T_{c0} = 1.70$ °K, we have $w = 8.63\times 10^{-6}$. With such small value of the Ginzburg critical width a negligible correction to the curve (a) in Fig. 2 is expected.

We exhibit the experimental results of the Al – 6 junction in the Fig. 3. We can see that using the GR model [Eqs. (2.11) and (2.12)], we get a very good fit to the experimental data. It is also seen that the HR-model and the GR-model results are almost the same for this particular set of data ($w \sim 10^{-5}$). In the insert of the Fig. 3, we show the curves of $\frac{1}{2}\Gamma(T)$ calculated from those two models in the very vicinity of T_c(1.7732 °K) on an enlarged scale.

Therefore, we conclude that the GR model can be used as a satisfactory explanation for this particular experiment[14] and all other possible experimental situations with such small values of w. However, measurements on films with larger values of w should, according to Fig. 2, exhibit a gradual changeover from the GR regime (curves

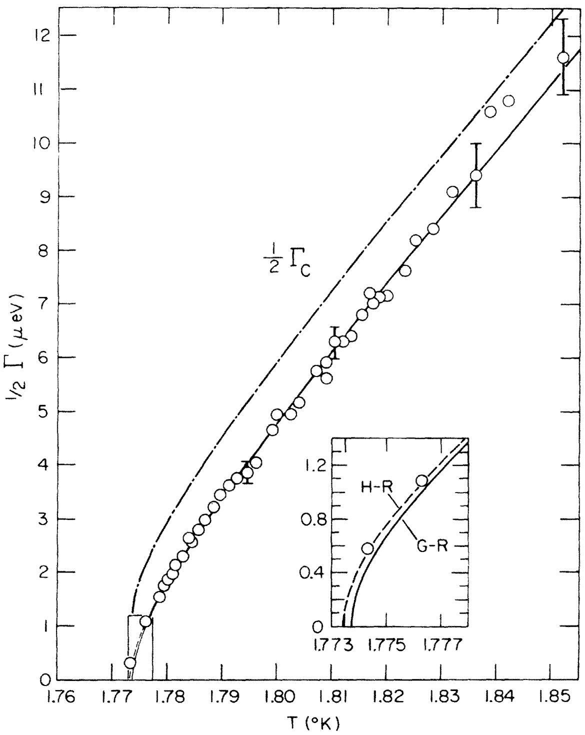

FIG. 3. Relaxation rate as a function of the absolute temperature T. The full (interrupted) line represents the critical (conventional) slowing-down calculated in GR model. Insert: The HR- and GR-model results in an expanded scale. The experimental data are for the Al-6 junction of the Ref. 4.

with a pronounced knee) towards a regime with rather different nonlinear shape.

Finally, we would like to point out that the present approach is not restricted to superconductors. It can be applied to random magnets[1] where the critical dynamics can be investigated using neutron-diffraction methods. An important implication of our present work is a prediction of a narrow central peak in the dynamic form factor owing to the presence of two different time constants in the relaxation function. This is demonstrated in Fig. 3 by the curve Γ_c, which represents the conventional slowing down, which very near T_c is much larger than the values corresponding to the critical slowing down. This feature suggests that the present calculation should be extended to three dimensions, in order to describe the dynamics of a magnetic glass. Such a calculation is presently underway, and will be published in a forthcoming paper.

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[1]D. Shapero, J. R. Cullen, and E. Callen, Phys. Lett. A 50, 303 (1974).

[2]A. J. Bray, Phys. Rev. B 13, 3807 (1976); A. I. Larkin, Zh. Eksp. Teor. Fiz. 58, 1466 (1970) [Sov. Phys.–JETP 31, 784 (1970)].

[3]J. C. Hayward and E. Šimánek, Phys. Lett. A 53, 55 (1975).

[4]J. T. Anderson, R. V. Carlson, and A. M. Goldman, J. Low Temp. Phys. 8, 29 (1972).

[5]K. Yoshihiro, K. Yamaji, and K. Kajimura, J. Phys. Soc. Jpn. 33, 100 (1972).

[6]I. Schüller and K. E. Gray, Phys. Rev. Lett. 36, 429 (1976).

[7]L. van Hove, Phys. Rev. 95, 1374 (1954).

[8]G. Rickayzen and A. J. Bray, J. Phys. F 3, L134 (1973).

[9]The parameter w is related to the Ginzburg critical width $\Delta\epsilon_G$ as $\Delta\epsilon_G = 8w^2$. See, e.g., L. P. Kadanoff, W. Götze, D. Hamblen, R. Hecht, E. A. S. Lewis, V. V. Palaciauskas, M. Rayl, and J. Swift, Rev. Mod. Phys. 39, 395 (1967).

[10]$E_1(z)$ is the exponent-integral function. See, e.g., M. Abramovitz and J. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1965), Chap. 5.

[11]M. Suzuki and G. Igarashi, Prog. Theor. Phys. 49, 1073 (1973).

[12]It is interesting to note that the same value was obtained for the specific-heat exponent α in the same (GR) approximation. E. Šimánek, J. Phys. F (to be published).

[13]The values of the parameters r_1 and r_2 were chosen in such a way as to fit the experiment described below.

[14]P. Walker and E. Šimánek, Physica B 82, 313 (1976).