

## Critical Fluctuations in the Thermodynamics of Quasi-Two-Dimensional Type-II Superconductors

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Thermodynamic quantities in quasi-2D type-II superconductors exhibit characteristic scaling behavior for high fields in the critical region around  $H_{c2}(T)$ . Using a nonperturbative approach to the Ginzburg-Landau free energy functional, the scaling functions for the free energy, magnetization, entropy, and specific heat are evaluated in a closed form. The experimental data for  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  are presented which are in agreement with the theoretical results.

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The properties of type-II systems have recently been under intense study, particularly in connection with high-temperature superconductors (HTS). A fundamental problem in this field is that of critical behavior arising from thermal fluctuations. Several experiments point to the importance of fluctuations in the thermodynamics of HTS [1-6]. In a recent experiment, Welp *et al.* [3] observed that the superconducting contribution to the magnetization and resistivity of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  crystals displays three-dimensional (3D) scaling behavior in the variable  $[T - T_c(H)]/(TH)^{2/3}$  around the critical temperature in the general vicinity of the upper critical field  $H_{c2}(T)$ . Li and Suenaga [6] noticed that the magnetization of highly anisotropic  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  crystals near critical temperature can be described by the 2D version of the scaling function in the variable  $[T - T_c(H)]/(TH)^{1/2}$ . The scaling indicates that the problem of fluctuations near  $H_{c2}(T)$  can be represented in terms of the Ginzburg-Landau (GL) field theory on a degenerate manifold spanned by the lowest Landau level (LLL) for Cooper pairs (we call it the GL-LLL theory). The GL-LLL description of fluctuations near  $H_{c2}(T)$  is formally valid if the  $(H, T)$  point lies above the  $\tilde{H}(T)$  line given by  $\tilde{H}(T) = (1/3)H_{c2}(T) + (\sqrt{\theta}/3)[\dot{H}(T)H_{c2}(0)T/T_{c0}]^{1/2}$ , where  $\theta \ll 1$  is the Ginzburg fluctuation parameter (see below). Below  $\tilde{H}(T)$  the interaction term in the GL theory is larger than the cyclotron gap of Cooper pairs and the fluctuations from excited Landau levels become significant. The GL-LLL description is valid everywhere in the critical region around  $H_{c2}(T)$  except for the area of size  $\sim \theta \ll 1$  surrounding  $[H = 0, T = T_{c0}]$ .

The scaling property of GL-LLL theory for a quasi-2D superconductor implies that the free energy  $F(T, H)$  near  $H_{c2}(T)$  must be of the form  $F(T, H) = THf(At)$ , where  $f(At)$  is a scaling function of variable  $t = [T - T_c(H)]/(TH)^{1/2}$  and  $A$  is a constant [7]. The function  $f(x)$  is known only in the limit  $x \gg 1$ , where perturbation theory can be used to account for the fluctuation contribution to the free energy. Various extrapolation

schemes have been used in the past to reconstruct the form of  $f(x)$  outside the perturbative regime ( $x \gg 1$ ), and, in particular, in the crossover region around  $x = 0$  [ $H_{c2}(T)$  line]. These schemes include the diagrammatic approach, [8] Padé and Borel-Padé approximants to the perturbation series [9], and possible connection with a simple 0D GL theory in zero field. Recently, a nonperturbative approach to this problem has been developed in Ref. [10]. In this Letter we use this approach to solve for the thermodynamics of quasi 2D type-II superconductors in the vortex phase and derive an explicit form for  $f(x)$ . After introducing the correct collective variables, the overall amplitude of the order parameter  $\Psi(\mathbf{r})$  and positions of vortices, we proceed to perform the integration over the overall amplitude exactly [10]. As a result, the correlations among vortices affect thermodynamics through interaction which can be thought of as an analog of the Abrikosov parameter  $\beta_A$  for arbitrary configurations of vortices [10]. Such interaction depends only weakly on vortex configurations, an example being the well-known small difference between  $\beta_A$  for a triangular and a square lattice. Neglecting the dependence of the generalized Abrikosov parameter on vortex correlations we find *explicit* closed form expressions for the scaling functions  $f(x)$  for free energy, as well as for magnetization, entropy, and specific heat. Our theoretical results are in very good agreement with the experimental magnetization data for  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  obtained in the high-field regime where the GL-LLL description and corresponding scaling behavior are valid [5, 6].

For our purposes it suffices to study a 2D system. The description of fluctuations in superconductors with weak Josephson coupling between the layers is based on the 2D GL functional at all temperatures except in a narrow interval near  $T_c$ , where the fluctuations have a 3D character. This temperature interval of 3D fluctuations  $\Delta T$  can be roughly estimated by comparison of the Josephson coupling energy with the intralayer condensation energy. This gives the condition  $\xi_c(T) \approx s$  where  $\xi_c(T)$  is the

correlation length along the  $c$  axis (perpendicular to the layers) and  $s$  is the effective interlayer spacing. One obtains  $\Delta T \approx T_c \xi_{ab}^2(0)/s^2 \gamma^2$  where  $\xi_{ab}(0)$  is the correlation length along the layers (in the  $ab$  plane) extrapolated to zero temperature, and  $\gamma$  is the anisotropy ratio. For Bi-based superconductors with anisotropy  $\gamma$  as high as 55 in the case of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  and 31 in  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  [11] with  $\xi_{ab}(0) \approx 20 \text{ \AA}$  and  $s \approx 20 \text{ \AA}$ , the region of 2D-like behavior of fluctuations covers practically all temperatures of interest.

We consider the limit  $\kappa \gg 1$  and ignore the fluctuations of the magnetic field (this is an excellent approximation in HTS where  $\kappa \sim 10^2$ ). The essential features of the critical behavior are then described by the partition function

$$Z = \int_{\mathcal{H}_0} \mathcal{D}\Psi(\mathbf{r}) \exp \left[ \frac{s}{T} \int dr \left\{ -\tilde{a} |\Psi(\mathbf{r})|^2 - \frac{1}{2} b |\Psi(\mathbf{r})|^4 \right\} \right], \quad (1)$$

where  $\tilde{a} = a(T)[1 - H/H_{c2}(T)]$ ,  $a$  and  $b$  are the GL coefficients,  $a(T) = a'(T - T_{c0})$ , and the functional integral is to be taken over the subspace  $\mathcal{H}_0$  spanned by the LLL. It is supposed that higher Landau levels are taken into account by renormalization of the GL parameters  $\tilde{a}(T)$  and  $b$ , and these parameters differ from the original ones for the GL model for  $H = 0$  due to contribution of higher

Landau levels. For  $H > \tilde{H}(T)$  this contribution is small, of order  $\theta^2$  and  $\theta$  for  $\tilde{a}(T)$  and  $b$ , respectively.

The confinement to the LLL eliminates gradients in the plane perpendicular to  $\mathbf{H}$ . This leads to the enhancement of fluctuations near  $H_{c2}(T)$  through a dimensional reduction [12]: At the perturbative level the fluctuations appear zero dimensional. The critical behavior of the GL-LLL theory (1), however, is deeply nonperturbative due to the constraint  $\Psi(\mathbf{r}) \in \mathcal{H}_0$ . To study this behavior we use the symmetric gauge to write  $\Psi(\mathbf{r})$  in LLL subspace as [10, 13]

$$\Psi(\mathbf{r}) = \Phi \prod_i^N (z - z_i) \exp(-|z|^2/4), \quad (2)$$

where  $z = (x + iy)/l$ ,  $l^2 = \phi_0/2\pi H$ ,  $N = SH/\phi_0$  is the number of vortices, and  $S$  is the total layer area. Variables  $\{z_i\}$  (positions of vortices) and  $\Phi$  (overall amplitude) are the correct collective modes of the GL-LLL theory (1). The partition function in Eq. (1) can now be written in terms of these new modes. The crucial point is that the integration over  $\Phi$  in Eq. (1) can be carried out exactly in the thermodynamic limit  $N \rightarrow \infty$  [10]. This recovers the crossover to the low- $T$  saddle point which controls the thermodynamics in the critical region and which is beyond the reach of perturbative expansions. The integral in question is

$$\int_0^\infty d|\Phi|^2 |\Phi|^{2N} \exp(-2\alpha|\Phi|^2 - \frac{1}{2}|\Phi|^4) \propto \sqrt{N!} \exp[-\alpha^2 - \alpha\sqrt{\alpha^2 + N} - N \sinh^{-1}(\alpha/\sqrt{N})], \quad (3)$$

where  $\alpha^2 + N \gg 1$  is assumed. Note that  $|\Phi|^{2N}$  arises from the Jacobian of the transformation implicit in Eq. (2) [10]. This finally leads to the partition function

$$Z \propto \frac{1}{N!} \int \prod_i^N \frac{dz_i dz_i^*}{2\pi} (\bar{f}^4)^{-N/2} \prod_{i < j}^N |z_i - z_j|^2 \exp[\frac{1}{2}NV^2 - \frac{1}{2}NV\sqrt{V^2 + 2} - N \sinh^{-1}(V/\sqrt{2})], \quad (4)$$

where  $V(\{z_i\}) = gU(\{z_i\})$  and

$$\bar{f}^p(\{z_i\}) = \int \frac{dz dz^*}{2\pi N} \exp(-p|z|^2/4) \prod_i |z - z_i|^p, \quad U(\{z_i\}) = \bar{f}^2/(\bar{f}^4)^{1/2}, \quad g = ta'(\phi_0 s/2b)^{1/2}. \quad (5)$$

The partition function (4) describes the thermodynamics of dense classical vortices. The important point is that the field and temperature dependence enter through the coupling constant  $g$  in front of the multiple-body configuration interaction  $U(\{z_i\}) = \bar{f}^2/(\bar{f}^4)^{1/2}$  only. This vortex interaction  $U$  is simply  $1/\sqrt{\beta_A}$ , but with Abrikosov parameter  $\beta_A(\{z_i\})$  evaluated for arbitrary configurations of vortices.

Now we use the fact that major rearrangements of vortices lead only to rather small changes in  $\beta_A$ . For example, as is well known, the difference in  $\beta_A$  for a triangular and (locally unstable) square lattice is only a few percent. Only collapsed or exploded zeros, which are highly improbable configurations, can change  $\beta_A$  and  $U$  significantly. This is confirmed by evaluating the thermody-

amic average  $\langle U \rangle$  in the perturbative limit  $g \gg 1$  where one expects  $\langle U \rangle$  to be smallest. The result is  $\langle U \rangle = 1/\sqrt{2}$  in this limit. Thus, over the full range of  $g$ ,  $\langle U \rangle$  changes from 0.707 to 0.928, the latter corresponding to the triangular lattice ( $g \rightarrow -\infty$  limit). We therefore expect that simply replacing  $U$  in Eq. (5) by a  $z_i$ -independent value  $U_0 (\cong \langle U \rangle)$  should be a very good approximation for all temperatures and fields where the scaling itself is valid. The essential physics is that the variation in thermodynamic functions through the critical region is caused primarily by the rapid change in  $|\Phi|^2$ ; In our nonperturbative approach this part of the problem is treated exactly. The change in  $U$  is comparatively very slow and is quantitatively less important.  $U_0$  should be chosen to

interpolate smoothly between the two limits: Here we simply use  $U_0 = 0.9$  far below  $H_{c2}(T)$ ,  $U_0 \sim 0.8$  around  $H_{c2}(T)$ , and  $U_0 = 0.7$  far above  $H_{c2}(T)$  [14]. After setting  $U = U_0$  the  $t$ -dependent part of  $Z$  can be found easily. Using this approximation we finally obtain the scaling functions for free energy density, magnetization, entropy density, and specific heat:

$$\frac{F(T, H)}{TH} s\phi_0 = f(x), \quad x = At, \quad A = a'(\phi_0 s/2b)^{1/2} U_0,$$

$$f(x) = -\frac{1}{2}x^2 + \frac{1}{2}x\sqrt{x^2 + 2} + \sinh^{-1}(x/\sqrt{2}), \quad (6)$$

$$\frac{M(H, T)}{\sqrt{HT}} \frac{s\phi_0 H'_{c2}}{A} = -f'(x) = x - \sqrt{x^2 + 2}, \quad (7)$$

$$\frac{\sigma(H, T)}{\sqrt{HT}} \frac{\phi_0}{A} = x - \sqrt{x^2 + 2}, \quad (8)$$

$$C(H, T) \frac{s\phi_0}{TA^2} = f''(x) = 1 - \frac{x}{\sqrt{x^2 + 2}}, \quad (9)$$

where  $H'_{c2} = |dH_{c2}/dT|$  at  $T = T_{c0}$ . In deriving (7), (8), and (9) from the free energy in Eq. (6) we have kept only the leading derivatives. It is evident from the form of scaling functions in Eqs. (6)–(9) that the critical behavior of the GL-LLL model in 2D is *different* from the 0D GL model [15].

The comparison to experiments indicates that the present form for  $f(x)$  is quite accurate (see below). This approach, however, describes only the “smooth” thermodynamics and cannot be used to extract information about the liquid-solid phase transition which was found to take place at  $g = g_M \sim -7.5$  [10]: For this transition the dependence of  $U$  on  $z_i$  is crucial. We note that it is this weak dependence of the generalized  $\beta_A$  on  $z_i$  which is responsible for the absence of a strong phase transition at the melting point (this is in agreement with experimental observations: Up to now singular behavior at the melting point was not observed in any thermodynamic quantity).

The thermodynamic functions for magnetization and entropy given in Eqs.(6)–(9) have the following remarkable property: Both  $M(H, T)$  and  $\sigma(H, T)$  are independent of  $H$  at temperature  $T^*$ , i.e., all  $M(T)$  or  $\sigma(T)$  curves for different  $H$  cross at the point  $T^*$ :

$$\frac{T_{c0} - T^*}{T^*} = \frac{bH'_{c2}}{a'^2 \phi_0 s U_0^2}. \quad (10)$$

We note that both the scaling form of magnetization (or entropy) and the existence of the crossing point (10) naturally lead to the function  $f'(x) = -x + \sqrt{x^2 + 2}$ . Thus the existence of the crossing point in the scaling regime is a direct consequence of the weak dependence of the generalized Abrikosov parameter  $\beta_A$  (i.e., the vortex interaction  $U$ ) on coordinates of vortices. The crossing point  $T^*$  lies in the region of strong (critical) fluctuations, the right-hand side of Eq. (10) being the Ginzburg fluctuation parameter  $\theta$  for quasi-2D superconductors.

The magnetization at the crossing point is

$$M(T^*) = T^*/s\phi_0. \quad (11)$$

Thus the ratio  $M(T^*)/T^*$  gives direct information on the effective interlayer spacing  $s$ . We note that this parameter coincides with the distance between layers given by the crystal structure in compounds with one superconducting layer per unit cell. In Bi-based superconductors there are two or three  $\text{CuO}_2$  layers per effective unit cell and only in the case of their strong superconducting coupling (in comparison with the intralayer condensation energy) they can be treated as one layer and the parameter  $s$  coincides with the size of the effective unit cell in the  $c$  direction. Otherwise the layers should be treated individually and the effective spacing  $s$  is given by the average distance between  $\text{CuO}_2$  layers.

Now we point out that the scaling approach describes the crossing point for fields

$$H \gtrsim \tilde{H}(T^*) \gtrsim \frac{1}{3}H^*, \quad H^* = H_{c2}(T^*) = H'_{c2}(T_{c0} - T^*). \quad (12)$$

For lower fields higher Landau levels are important. In the low-field regime  $H \ll H_{c2}$  (in the London region) the contribution of the vortex fluctuations to the thermodynamics was described in [16]. In the London region the amplitude of the order parameter is constant while positions of vortices  $z_i$  fluctuate. The  $H$  dependence of the magnetization is logarithmic in this region. The slope  $dM/d \ln H$  depends on  $T$  and becomes zero at  $T = T^*$ , where  $T^*$  is again given by Eq. (10), but now with  $U_0^2$  replaced by  $1/2$ . Thus the crossing point  $T^*$  in  $M$  vs  $T$  curves at different fields  $H$  exists in the London region of the vortex state as well, i.e., for  $H \ll H^*$ . Noting that such a crossing is absent in the mean-field approach, we conclude that the existence of the crossing point  $T^*$  is a general consequence of fluctuations in the vortex state. Formation of the crossing point is caused by the entropy associated with fluctuations of vortices; in the low-field regime the fluctuations of vortex positions are important while in the high-field regime the amplitude fluctuations give the main contribution.

We now use our results to understand the experimental data on magnetization for highly  $c$ -axis oriented  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  with the magnetic field parallel to the  $c$  axis [5]. In the scaling regime we use the expression

$$\frac{M}{M^*} = \frac{1}{2} [1 - \tau - h + \sqrt{(1 - \tau - h)^2 + 4h}], \quad (13)$$

$$\tau = (T - T^*)/(T_{c0} - T^*), \quad h = H/H^*,$$

which follows from Eq. (7). The values  $T^* = 108.1$  K and  $4\pi M^* = -2.67$  G were taken directly from the data for the crossing point. Two other parameters  $T_{c0}$  and  $H'_{c2}$  were obtained by the least-squares minimization method using Eqs. (13) and the experimental data for temperatures between 107 and 120 K and fields  $5 \geq H \geq 1$  T where the scaling approach works well. From this fit

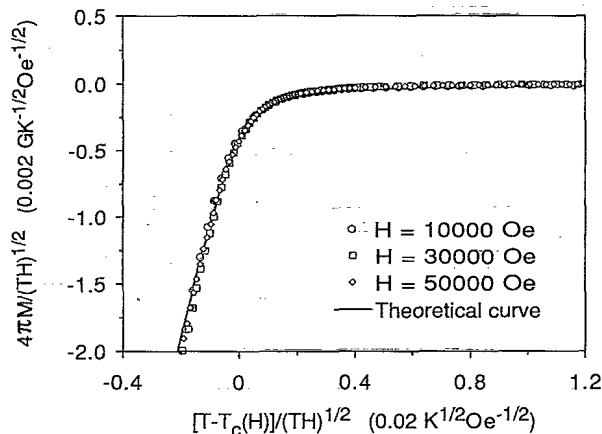


FIG. 1. 2D scaling of magnetization data for  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  in various magnetic fields parallel to the  $c$  axis. The theoretical curve is obtained from scaling function  $f'(x)$  given by Eq. (7).

we obtain  $T_{c0} = 111$  K and  $H'_{c2} = 3.44$  T/K, as well as  $\theta = 0.027$ . The theoretical curve given by Eq. (7) and experimental data for different temperatures and fields are shown in Fig. 1. Note that fitting parameter  $H'_{c2}$ , as well as parameters  $b/a'^2$  and  $T_{c0}$  in (10) obtained in the high-field regime may differ somewhat from the corresponding parameters of the original GL functional at  $\mathbf{H} = 0$  due to the contribution of higher Landau levels mentioned above.

In summary, starting from the Ginzburg-Landau theory we have presented the universal scaling functions for the thermodynamic quantities in quasi-2D type-II superconductors. These functions, as well as the theoretical conclusion about existence of the crossing point for  $M(T)$  curves at different fields within the scaling region, are in accordance with experimental data.

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