Density-Functional Theory of Freezing of Vortex Liquid in Quasi-Two-Dimensional Superconductors

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We present a theory of the vortex-liquid-to-solid transition in homogeneous quasi-2D superconductors. The free energy is written as a functional of the density of zeros of the fluctuating order parameter. The transition is weakly first order and well below the $H_{c2}(T)$ line. Transition temperature, discontinuities of the average Abrikosov ratio and of the average Cooper pair density, the Debye-Waller factor, and the latent heat are in good agreement with Monte Carlo simulations. The density is only weakly modulated in the "vortex-solid" phase, consistent with the density-wave behavior.

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The experimental observation [1] of the wide region around the $H_{c2}(T)$ line where the Abrikosov lattice appears melted demonstrates the importance of thermal fluctuations in high-temperature superconductors (HTS). Such fluctuations are generally important in quasi-two-dimensional (2D) materials, such as thin films or layered systems with weak interlayer coupling. The existence and the nature of the ordered low-temperature phase are still matter of some controversy. The early theories [2] *assumed* the ordered phase at low temperatures and proposed dislocation unbinding as a mechanism of second-order melting transition or used the Lindemman criterion to locate the melting point [3]. However, even the very existence of the vortex-lattice phase, which is the starting point of these theories, cannot be taken for granted [4, 5]. External magnetic field causes the system to appear $D - 2$ dimensional on the perturbative level, and the perturbation theory carried out to the eleventh order [6] shows no sign of the low-temperature Abrikosov phase in 2D. Renormalization group analysis near the upper critical dimension $D = 6$ found the continuum of relevant charges to be generated by the renormalization procedure and indicated that the transition into the Abrikosov vortex-lattice state might be first order, as expected from Landau argument [7]. The relevance of this analysis for real superconductors, however, is not obvious since the lower critical dimension for the Abrikosov transition is $D = 4$ [5]. Recent Monte Carlo (MC) simulations [8–10] revealed a weak first-order vortex-liquid-to-solid transition in 2D. At present, our information about this transition comes almost exclusively from such numerical work.

The main purpose of this paper is to develop a working *analytic* theory which has predictive power and which can be applied to a variety of experimentally relevant issues. We propose a density-functional representation of the free energy for zeros of the fluctuating superconducting order parameter, with the exact two-body correlations built in. Our theory describes *strong fluctuations,* in the sense that it does not rely on the expansion around the Abrikosov lattice. In fact, it is an expansion around the *liquid* state, allowing us to study non-Abrikosov phases [5]. It is shown that above a certain value of the vortex liquid structure factor the triangular lattice has lower free energy than the uniform configuration. We calculate several typical quantities like the latent heat and the change in thermal average of Abrikosov's ratio at the transition and use the information on the dependence of the structure factor on temperature from the existing MC simulations to locate the transition temperature. Obtained results are in good agreement with those found in numerical simulations.

The starting point is the Ginzburg-Landau (GL) partition function for a homogeneous 2D superconductor in perpendicular magnetic field, with fluctuations of the magnetic field neglected ($\kappa \gg 1$). We are interested in the regime where the Landau level (LL) structure of Cooper pairs dominates the fluctuation spectrum: This is the case for fields above $H_b \approx (\theta/16)H_{c2}(0)(T/T_{c0})$, where θ is the Ginzburg fluctuation parameter [11]. (For example, in BiSrCaCuO 2:2:1:2, $\theta \sim 0.045$ and $H_b \sim 1$ T.) In this regime, the approximation in which one retains only the lowest Landau level (LLL) modes describes essential features of the problem. The partition function is then $Z = \int D[\psi^*\psi]\exp(-S)$, and

$$S = \frac{d}{T}\left\{\alpha'(T)\int d^2\mathbf{r}\,|\psi(\mathbf{r})|^2 + \frac{\beta}{2}\int d^2\mathbf{r}\,|\psi(\mathbf{r})|^4\right\}, \tag{1}$$

where $\alpha'(T) = \alpha(T)[1 - H/H_{c2}(T)]$, d is the thickness of the film or the effective interlayer separation, and $\alpha(T)$ and β are phenomenological parameters. We choose to work in the symmetric gauge so that the order parameter is a holomorphic function and can be written as $\psi(z) = \phi\prod_{i=1}^{N}(z - z_i)\exp[-(|z|^2/4)]$ where N is the area of the system in units of $2\pi l^2$, $z = (x + iy)/l$, and l is the magnetic length for the charge $2e$. The partition function can now be expressed in terms of variables ϕ and $\{z_i\}$ [8]. They represent two distinct tendencies in a superconductor; ϕ describes the overall growth of the local superconducting order, while $\{z_i\}$

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represent the remaining weak lateral correlations between vortices. The integration over ϕ can be performed exactly in the thermodynamic limit $N \to \infty$, yielding $Z = \int \prod_i (dz_i\, dz_i^*/2\pi)\, \bar{f}^{4-N/2} \prod |z_i - z_j|^2 \exp\{-S'\}$, where

$$S' = -\frac{V^2}{2} + \frac{V}{2}(V^2+2)^{1/2} + \sinh^{-1}\left(V/\sqrt{2}\right), \tag{2}$$

$V(\{z_i\}) = g\bar{f}^2/\sqrt{\bar{f}^4}$, $\bar{f}^n(\{z_i\}) = \int (dz\, dz^*/2\pi N) \prod_i |z - z_i|^n \times \exp[-(n|z|^2/4)]$, and $g = \alpha' \sqrt{\pi l^2 d/\beta T}$. Variable ϕ is determined by

$$\langle|\phi|^2\rangle \bar{f}^2 \frac{2\pi l^2 d|\alpha'|}{T} = V^2(1 + \sqrt{1 + 2V^{-2}}). \tag{3}$$

The original problem is now equivalent to the thermodynamics of classical 2D system of particles which we call dense-vortex plasma (DVP) [8]. We note two important features of the DVP system: (1) Particles interact via long-range multiple-body forces causing the system to be incompressible, and (2) the system is scale invariant so that the thermodynamics depends on a single dimensionless coupling constant g. Previous MC simulations show the transition from a liquid to a solid state taking place at $g_M \approx -7$ [8–10]. From Eq. (2) we can write the *exact* free energy per vortex as $f(g)/T = S' - s(\langle\beta_A\rangle)$ where $\langle\beta_A\rangle$ is the g-dependent thermal average of the Abrikosov ratio $\beta_A(\{z_i\}) = \bar{f}^4/(\bar{f}^2)^2$, determined by minimizing $f(g)$. The entropy $s(\langle\beta_A\rangle)$ is given by

$$s(\langle\beta_A\rangle) = N^{-1} \ln \int \prod_i \frac{dz_i\, dz_i^*}{2\pi l^2} \bar{f}^{4-N/2} \times \prod_{i<j} |z_i - z_j|^2 \delta\left(\frac{\bar{f}^4}{(\bar{f}^2)^2} - \langle\beta_A\rangle\right). \tag{4}$$

The "smooth" part of $s(\langle\beta\rangle)$ (4) can be evaluated by using the "98%–2%" procedure of [11]. Here we concentrate on the 2% piece of the entropy carrying the information about phase transitions.

To study the solidification transition it is beneficial to change variables from particle coordinates to density of particles. To achieve this we insert $1 = \int D\rho(\mathbf{r})\delta[\rho(\mathbf{r}) - \sum \delta(\mathbf{r} - \mathbf{r}_i)]$ in Z (2) [12]. After the δ function is expressed as an integral over auxiliary field $\Phi(\mathbf{r})$, the coordinates of vortices are integrated out. The partition function becomes $Z = \int D\rho(\mathbf{r}) D\Phi(\mathbf{r}) \exp\{-S''\}$,

$$S'' = U[\rho(\mathbf{r})] - \int d^2\mathbf{r}\Phi(\mathbf{r})\rho(\mathbf{r}) - N \ln \int d^2\mathbf{r} \exp[-\Phi(\mathbf{r})], \tag{5}$$

where $U[\rho]$ is the exact energy density functional determined by the details of the DVP interaction. The full form of $U[\rho]$ is not important for the present purposes; we expand U in terms of multibody interactions and keep only the first, two-body term so that $U \approx \frac{1}{2}\int \rho(\mathbf{r}_1) v(\mathbf{r}_1 - \mathbf{r}_2)\rho(\mathbf{r}_2) + O(\rho^3)$. After this truncation of the energy functional, density is integrated out, leaving the partition function as an integral over field $\Phi(\mathbf{r})$ with the action

$$\frac{F}{T} = -\frac{1}{2}\int d\mathbf{r}_1\, d\mathbf{r}_2 \Phi(\mathbf{r}_1) v^{-1}(\mathbf{r}_1 - \mathbf{r}_2)\Phi(\mathbf{r}_2) - N \ln \int d^2\mathbf{r} \exp[-\Phi(\mathbf{r})]. \tag{6}$$

We take Eq. (6) *to be the desired expression for the mean-field free energy of the vortex system.* Under this assumption it follows that $-v(r) = c(r)$, where $c(r)$ is the direct (Ornstein-Zernike) correlation function of the vortex liquid whose Fourier transform is related to the structure factor via the relation $S(q) = [1 - c(q)]^{-1}$ [13]. The structure factor $S(q)$ is the Fourier transform of the connected density-density correlation function.

Before proceeding with the analysis of the phases described by the free energy (6) let us comment on the validity of the truncation of the DVP energy density functional. Even if the particles interact only via two-body forces all multibody correlations will be generated and the true free energy would contain additional terms of higher order in field $\Phi(\mathbf{r})$. Therefore, the fact that vortices in DVP interact through multibody forces makes the approximate free energy (6) no less appropriate here than for the ordinary correlated liquid. In fact, one might expect that the long-range interaction operating in DVP will make the system only more mean-field-like. The free energy (6) may be thought of as a harmonic approximation around the liquid state and this particular form is appropriate for an incompressible system [13].

We assume that DVP freezes into a triangular lattice with period a, $a^2 = 4\pi/\sqrt{3}$ and we work in units $l = 1$. Note that this assumption is not necessary and that we could consider other lattices. Actually, at this point it is not obvious that the ordering wave vector will be in any simple relationship to the magnetic length. Within the resolution of the MC data however, the strongest tendency for ordering corresponds to the triangular modulation. We determine the solid phase by two real order parameters Φ_1 and Φ_2 in the following way: $\Phi(\mathbf{r}) = \Phi_0 - 2\sum_{i=1}^{2} \Phi_i f_i(\mathbf{r})$, where $f_1(\mathbf{r}) = (1/2)\sum_{G_1} \exp[(i\mathbf{G}_1)\cdot\mathbf{r}] = \cos(ax) + 2\cos(a\sqrt{3}y/2) \times \cos(ax/2)$ and the sum runs over six shortest reciprocal lattice vectors $|\mathbf{G}| = G_1 = a$. The same sum, but over next six shortest reciprocal vectors $|\mathbf{G}| = G_2 = a\sqrt{3}$ determines the function $f_2(\mathbf{r}) = \cos(a\sqrt{3}y) + 2\cos(3ax/2) \times \cos(a\sqrt{3}y/2)$. Inserting this into expression (6) we get two-order-parameter form of

the free energy per vortex,

$$\frac{F}{T} = \frac{\Phi_0^2}{2c_0} + \Phi_0 + 3\sum_{i=1}^{2}\frac{\Phi_i^2}{c_i} - \ln\int d^2\mathbf{r}\exp\left(2\sum_{i=1}^{2}\Phi_i f_i(\mathbf{r})\right), \quad (7)$$

where $c_{1/2} = c(G_{1/2})$. The free energy is minimized by the solutions of the equations:

$$\frac{3\Phi_j}{c_j} = \frac{\int d^2\mathbf{r} f_j(\mathbf{r})\exp\left[2\sum_{i=1}^{2}\Phi_i f_i(\mathbf{r})\right]}{\int d^2\mathbf{r}\exp\left[2\sum_{i=1}^{2}\Phi_i f_i(\mathbf{r})\right]} \quad (8)$$

for $j = 1, 2$. The equation for the uniform component is $\Phi_0 = -c_0$, which means that the average density $\rho(\mathbf{G} = 0) = 1$ does not change. It is easily seen that $\int f_j(\mathbf{r}) = 0$ and one solution of the equations is always $\Phi_1 = \Phi_2 = 0$ which describes the liquid state of DVP. To find a nonzero solution, we choose a pair (c_1, c_2), numerically solve Eqs. (8), and compare free energies of the solid and the liquid configurations. This way one obtains the crosses joined by the full line with a negative slope on the structural phase diagram presented on Fig. 1. For values of S_1 and S_2 above the line triangular lattice vortices is the stable phase, while below it DVP is in the liquid phase. The values of order parameters are approximately the same for all points on the transition line and they are $\Phi_1 = 0.50$ and $\Phi_2 = 0.10$.

To estimate the value of the coupling constant g_M where the DVP freezes the information on S_1 and S_2 as functions of g is needed. The vortex structure factor can be found by using the perturbation theory to determine the structure factor for $|\psi(\mathbf{r})|^2$, and then connecting the two through the expression for $\psi(\mathbf{r})$ given below Eq. (1) [14]. While this is possible in principle it is more expedient here to utilize the vortex structure factors from MC simulations [8, 15]. Particularly useful is a detailed analysis of the peaks in the vortex structure factor by O'Neill and Moore [15]. From MC simulations in spherical geometry they find that, for $5 < g^2 < 50$, both peaks depend approximately linearly on g^2. The straight line with positive slope in Fig. 1 represents the $S_1 - S_2$ relation derived from there. Two lines intersect at $S_1^M = 4.45$ and $S_2^M = 1.47$, which gives $g_m = -6.5$. Interestingly, around this value of the coupling constant O'Neill and Moore first start to see critical slowing down of the dynamics in their MC simulations, but they argued against finite temperature phase transition. Our result is in good agreement with MC simulations in Refs. [8–10].

The main features of DVP (long-range interactions, incompressibility) resemble those of another well studied classical system, namely, 2D one component Coulomb plasma (OCP) [16, 17]. It is therefore not surprising that the values of the first two peaks of the structure factor that we found at the transition closely match the numbers found in MC simulations there. In particular, the value of S_1^M is very close to 4.4 which is known as the 2D version of the Verlet criterion. For comparison, our number for S_2^M is much worse for the hard-disk system where it equals 1.9 [16]. This is because the system of hard disks is compressible and the proposed form of the mean-field free energy (6) ignores the change in average density at the freezing transition.

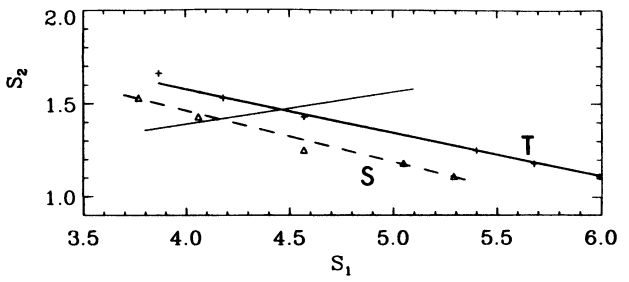

FIG. 1. Structural phase diagram for vortex system in terms of first two peaks of the structure factor of vortices. Crosses denote the values of S_1 and S_2 where the vortex solid becomes energetically favorable (transition line "T"), and triangles mark the points where the solid first becomes locally stable (spinodal line "S"). The lines through crosses and triangles are drawn to guide the eye. The line with positive slope depicts the $S_1 - S_2$ relation inferred from MC simulations [15]. The transition occurs at the intersection of this line with the transition line.

Since the transition takes place at a finite value of S_1 it is first order, in agreement with recent experiments in clean YBaCuO single crystals [18]. The latent heat comes solely from the structural change at the transition and it equals $Q = NT_M 3g_M^2\sum_{i=1}^{2}[\phi_i/(S_i^M - 1)]^2[dS_i^M/dg^2]$. This gives $Q/NT_M = 0.30$, close to 0.38 found in MC simulations [9, 10, 16]. Going back to the exact expression for the free energy of DVP [Eqs. (2) and (4)] it follows that the thermal average of Abrikosov ratio has a discontinuity at the transition $\Delta\langle\beta_A\rangle/\langle\beta_A\rangle \approx \langle\beta_A\rangle\Delta Q/g_M^2 NT_F = 0.01$ if we take $\langle\beta_A\rangle \approx 1.2$ at $g \approx g_M$. Invoking Eq. (3) we see that the spatially averaged $\langle|\psi(\mathbf{r})|^2\rangle$ has a discontinuous jump of about 1%. This agrees with the results of Refs. [9] and [10]. The Fourier components of the density of vortices can be determined from

$$\rho(\mathbf{G}) = \frac{\int d^2\mathbf{r}\exp[i\mathbf{G}\cdot\mathbf{r} - \Phi(\mathbf{r})]}{\int d^2\mathbf{r}\exp[-\Phi(\mathbf{r})]} \quad (9)$$

and the Debye-Waller factor is given by $\nu(G) = |\rho(G)|^2$. We find that for $|\mathbf{G}| \geq G_1$, $\rho(G) \approx 0.72\exp(-\lambda^2 G^2)$ with $\lambda = 0.47$. Similar Gaussian falloff was found for freezing into 3D bcc or fcc lattices, but with smaller coefficient λ ($\lambda_{\mathrm{bcc}} = 0.34$, $\lambda_{\mathrm{fcc}} = 0.19$ [13]). The fast decay of the Debye-Waller factor in DVP indicates that the transition is very weakly first order and the density modulation is rather small right below the transition.

The present scheme allows us to examine the possibility of the high temperature solid phase. We determined the values of (c_1, c_2) where the local $\Phi_1 \neq 0$, $\Phi_2 \neq 0$ minimum of the free energy first appears; at that point free energy of the liquid state is still lower than that of the solid. In this way the triangles joined by the dashed line on Fig. 1 are obtained. Intersection with the $S_1 - S_2$ line then determines the spinodal point: $g_{sh} = -6.25$. The interval $\Delta g_{sh} = g_{sh} - g_M = 0.25$ agrees well with the value of 0.2 obtained in Ref. [9] for the finite size system. The supercooling of the liquid phase is also possible, and the lowest $g = g_{sc}$ where the liquid phase is locally stable is given by the value where the first peak of $S(\mathbf{q})$ diverges. From the data in Ref. [9] we expect that the interval $\Delta g_{sc} = g_M - g_{sc}$ is of the same size as Δg_{sh}. The supercooling and the superheating data from the same reference indicate that there is a well defined critical value of the Abrikosov ratio $\langle \beta_A \rangle \approx 1.18$ at which the DVP system can undergo a continuous transition.

The vortex-liquid freezing transition discussed here is not a superconducting transition in the sense of breaking of U(1) symmetry [5]. It is best thought of as a transition to a charge-density wave of Cooper pairs. This is consistent with the weak density modulation of the ordered phase. The superconducting pairing susceptibility remains short ranged in this density wave, although its range becomes $\gg l$ as density modulation increases. This phase should be distinguished from other phases described in the literature [19]. A true superconducting transition in high magnetic field is almost always due to disorder, and, unless the disorder is strong, it will be much below the freezing transition discussed here [5]. Finally, the weak interlayer coupling in HTS and similar systems acts to "lock-in" density waves from different layers resulting in a 3D-ordered weakly modulated structure. The first order character of the transition will be enhanced. The change in the transition temperature will be rather small and can be readily estimated [8]. While we have used the numerical results as an input to our theory, it is not necessary to do so. Large-order perturbation expansion results for the structure factor of $|\psi(\mathbf{r})|^2$ [20] could have been used instead so that the theory is kept entirely at the analytic level.

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Note added.—Recently Sášik and Stroud [Phys. Rev. B **49**, 16074 (1994)] have evaluated numerically the discontinuity in the shear modulus, μ, at the transition. They find $\mu = 0.96$ in units in which $\mu_{KTBHNY} = \sqrt{3}$. Our theory gives $\mu = 1.6 \pm 0.7$. The error bounds reflect uncertainty in the curvature of the numerical structure factor at its first two peaks [15].

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