Density-functional theory of freezing of vortex-liquid in quasi two-dimensional superconductors

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Abstract: We present a theory of vortex liquid-to-solid transition in homogeneous quasi 2D superconductors. The free energy is written as a functional of density of zeroes 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 superfluid 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\cite{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 ordered low-temperature phase is still matter of some controversy. The early theories\cite{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\cite{3}. However, even the very existence of vortex-lattice phase, which is the starting point of these theories, cannot be taken for granted\cite{4,5}. External magnetic field causes the system to appear D-2 dimensional on perturbative level, and the perturbation theory carried out to eleventh order\cite{6} shows no sign of 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 Abrikosov vortex-lattice state might be first order, as expected from the simple Landau argument\cite{7}. The relevance of this analysis for real superconductors, however, is not obvious since the lower critical dimension for the Abrikosov transition is $D=4$. Recently, Monte Carlo (MC) simulations\cite{8,9,10} revealed very 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 wide variety of experimentally relevant issues. We derive a density-functional representation of the free energy for zeroes of the fluctuating superconducting order parameter, with the exact two-body correlations built in. Our theory is a “first principle” in the sense that it can be obtained from the microscopic description by a series of well understood approximations. It is then shown that above certain values of the first two peaks in the vortex structure factor triangular lattice has lower
free energy than the uniform configuration. Using the information on the dependence of the structure factor on temperature from the existing MC simulations we locate the transition temperature and calculate several typical quantities like the latent heat and the change in thermal average of the Abrikosov’s ratio at the transition. 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 $H_b \approx (\theta/16)H_{c2}(0)(T/T_c)$, where $\theta$ is the Ginzburg fluctuation parameter \[\text{[11]}\] (For example, in BSCCO 2:2:1:2, $\theta \sim 0.045$ and $H_b \sim 1$ Tesla). 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

$$S = \frac{d}{T}\{\alpha'(T) \int d^2\vec{r}|\psi(\vec{r})|^2 + \frac{\beta}{2} \int d^2\vec{r}|\psi(\vec{r})|^4 \},$$ (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 $\beta$ 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 $\phi$ and $\{z_i\}$ \[\text{[8]}\]. They represent two distinct tendencies in a superconductor; $\phi$ describes the overall growth of the local superconducting order, while $\{z_i\}$ represent the remaining weak lateral correlations between vortices. If the latter are treated in a certain average way one can account for 98% of the thermodynamics \[\text{[11]}\]. However, the transition that we want to discuss is entirely within the remaining 2%, and the correlations among $\{z_i\}$ are crucial. The integration over $\phi$ can be performed exactly in the thermodynamic limit $N \to \infty$, yielding $Z = \int \prod_i (dz_i dz_i^* / 2\pi) \bar{f}^{-N/2} \prod |z_i - z_j|^2 \exp \{-S'\}$,
where
\[ S' = -\frac{V^2}{2} + \frac{1}{V^2} (V^2 + 2)^{1/2} + \sinh^{-1}(V/\sqrt{2}) \],
\[ V([z_i]) = g\bar{f}^2/\sqrt{\bar{f}^4}, \bar{f}^2([z_i]) = f(dzd^*z/2\pi N) \prod_i |z-z_i|^n \exp -(n|z|^2/4) \text{ and } g = \alpha' \sqrt{\pi l^2/\beta T}. \]

The variable \( \phi \) is determined by
\[ <|\phi|^2 > f^2 \frac{2\pi l^2 d|\alpha'|}{T} = V^2 (1 + \sqrt{1 + 2V^2}). \]

The original problem is now equivalent to the thermodynamics of classical 2D system of particles which we call dense-vortex-plasma (DVP). We note several features of DVP system which should be reflected in our density-functional: 1) particles interact via long-range multiply-body forces, 2) the system is incompressible, 3) the system is scale invariant, 4) the thermodynamics depends on a single dimensionless coupling constant \( g \). Previous MC simulations show transition from a liquid to a solid state taking place at \( g_F \approx -7 \) [8, 9, 10]. The mechanism of the transition, however, remains obscure in the partition function (2) since the exponentiated energy is still a perfectly smooth function of \( V([z_i]) \). From Eq. (2) we can write the exact free energy per vortex as
\[ f(g)/T = S' - s(<\beta_A>) \] where \(<\beta_A>\) is the \( g \)-dependent thermal average of the Abrikosov ratio \( \beta_A([z_i]) = \bar{f}^4/\bar{f}^2 \) determined by minimizing \( f(g) \). The entropy \( s(<\beta_A>) \) is given by
\[ s(<\beta_A>) = N^{-1} \ln \int \prod_i \frac{dz_i z_i^*}{2\pi l^2} \bar{f}^4 - N/2 \prod_{i<j} |z_i - z_j|^2 \delta(\bar{f}^4/(\bar{f}^2)^2 - <\beta_A>). \]

The transition is (as usual) hidden in the entropy part of the free energy, but the evaluation of the integral (4) is a hopeless task. Obviously, a different route needs to be taken.

To study solidification transition it is often beneficial to change variables from particle coordinates to density of particles. This can be achieved by inserting the unity \[ 1 = \int D\rho(\bar{r}) \delta[\rho(\bar{r}) - \sum \delta(\bar{r} - \bar{r}_i)] \] in \( Z \) (2). After the \( \delta \)-function is expressed as an integral over auxiliary field \( \Phi(\bar{r}) \) coordinates of vortices can be integrated out. The partition function becomes
\[ Z = \int D\rho(\bar{r}) D\Phi(\bar{r}) \exp \{-S''\}, \]
\[ S'' = U[\rho(\bar{r})] - \int d^2\bar{r}\Phi(\bar{r})\rho(\bar{r}) - N \ln \int d^2\bar{r} \exp (-\Phi(\bar{r})) \]
where \( U[\rho] \) is the energy density-functional determined by the details of the DVP interaction and we changed variables \( i\Phi \rightarrow \Phi \). We expand \( U[\rho] \) in terms of multy-body interactions and keep only the first, two-body term so that \( U = \frac{1}{2} \int \rho(\vec{r}_1)v(\vec{r}_1 - \vec{r}_2)\rho(\vec{r}_2) + O(\rho^3) \). After this truncation of the energy functional, density can be integrated out leaving the partition function as an integral over field \( \Phi(\vec{r}) \) with the action

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

We take the Eq. (6) to be the desired expression for the mean-field free energy of the vortex system. In order to account for the strong two-particle correlations that exist in the liquid state of DVP we make the substitution \(-v(r) = c(r)\), 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 \( S(q) = (1 - c(q))^{-1} \). 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 validity of the truncation of the DVP energy density functional. Even if the particles interacted only via two-body forces all multi-body correlations will be generated and the true free energy would contain additional terms in (6) of higher order in field \( \Phi(\vec{x}) \). Therefore, the fact that particles in DVP interact through multi-body 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 important feature of the approximate free energy (6) is the last term which is purely entropic and hence crucial for the freezing transition. The free energy (6) may be thought of as a harmonic approximation around the liquid state and it will be shown that this particular form of it is very good for an incompressible system.

We assume that the freezing of the DVP happens into 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 for the structure factor, however, the strongest tendency for ordering corresponds to the triangular modulation. We determine the solid phase by two real order parameters $\Phi_1$ and $\Phi_2$ in the following way: $\Phi(\vec{r}) = \Phi_0 - 2\sum_{i=1}^{2} \Phi_i f_i(\vec{r})$, where $f_1(\vec{r}) = 1/2 \sum G_i \exp i\vec{G}_i \vec{r} = \cos(ax) + 2 \cos(a\sqrt{3}y/2) \cos(ax/2)$ and the sum runs over six shortest reciprocal lattice vectors $|\vec{G}| = G_1 = a$. The same sum, but over next six shortest reciprocal vectors $|\vec{G}| = G_2 = \sqrt{3}a$ determines the function $f_2(\vec{r}) = \cos(a\sqrt{3}y) + 2 \cos(3ax/2) \cos(a\sqrt{3}y/2)$. Inserting this into expression (6) we get two-order-parameter form of the free energy per vortex

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

(7)

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

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

(8)

for $j = 1, 2$. The equation for the uniform component is $\Phi_0 = -c_0$ and system is manifestly incompressible since the average density $\rho(\vec{G} = 0) = 1$ does not change with temperature. It is easily seen that $\int f_j(\vec{r}) = 0$ and one solution of the equations is always $\Phi_1 = \Phi_2 = 0$ which describes the liquid state of DVP. To find a non-zero 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 of vortices is the stable phase, while below it DVP is in liquid phase.

To find the value of the coupling constant $g_F$ 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 the superfluid density, $|\psi(\vec{r})|^2$, and then connecting the two through the expression for $\psi(\vec{r})$ given below Eq. (1) and some of the standard correlation function hierarchies (BBGKY, hypernetted chain, etc.). While
this is possible in principle it is more expedient here to utilize the vortex structure factors from MC simulations [8, 13]. Particularly useful is a detailed analysis of the peaks in the vortex structure factor by O’Neill and Moore [13]. In the MC simulations in the 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 $S_1 - S_2$ relation derived from there. Two lines intersect at $S_1^F = 4.45$ and $S_2^F = 1.47$, which gives $g_F = -6.5$ and the values of order parameters at the transition are $\Phi_1 = 0.50$ and $\Phi_2 = 0.10$. 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 excellent agreement with MC simulations in Refs. [8, 9, 10].

The main features of DVP (long-range interactions, incompressibility) resemble those of another well studied classical system, namely 2D one component Coulomb plasma [14]. It is therefore not surprising that the values of 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 $S_1^F$ is very close to 4.4 which is known as 2D version of the Verlet criterion. For comparison, our number for $S_2^F$ is much worse for the hard-disk system where it equals 1.9 [14]. 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.

Since the transition takes place at a finite value of $S_1$ the transition is first order in agreement with what has been observed in recent experiments in clean YBaCuO single crystals [15]. The latent heat comes solely from the structural change at the transition. If we take the expression for entropy of the system to be $s = \int d^2 \vec{r} \rho(\vec{r}) ln[\rho(\vec{r})]$ the latent heat equals $Q = NT_F 3(\Phi_1^2/c_1^F + \Phi_2^2/c_2^F)$. For the above numbers this gives $Q/NT_F = 1.0$, somewhat higher than the number 0.4 found in MC simulations [3, 9, 13]. Going back to the exact expression for the free energy it follows that the thermal average of Abrikosov ratio
has a discontinuity at the transition $\Delta < \beta_A > / < \beta_A > \approx < \beta_A > \Delta Q/g_F^2 NT_F = 0.025$ if we take $< \beta_A > \approx 1.2$ at $g = g_F$. Invoking Eq. (3) we see that the average superfluid density has a discontinuous jump of about 2%. This in reasonable agreement with the results of Refs. [9] and [10]. The Fourier components of the density of vortices can be determined from:

$$\rho(\vec{G}) = \frac{\int d^2\vec{r}\exp(i\vec{G}\cdot\vec{r} - \Phi(\vec{r}))}{\int d^2\vec{r}\exp(-\Phi(\vec{r}))}$$  \hspace{1cm} (9)$$

and the Debye-Waller factor is given by $\nu(G) = |\rho(G)|^2$. We find that for $|\vec{G}| \geq G_1 \rho(G) \approx 0.72 \exp(-\lambda^2 G^2)$ with $\lambda = 0.47$. Similar Gaussian fall-off was found for freezing into 3d bcc or fcc lattices, but with smaller coefficient $\lambda$ ($\lambda_{bcc} = 0.34$, $\lambda_{fcc} = 0.19$, [12]). The fast decay of Debye-Waller factor indicates that the transition is very weakly first order and the density modulation is rather weak right below the transition.

The present scheme allows us to examine the possibility of 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 the one of the solid. This way triangles joined by the dashed line on Fig. 1 are obtained. Intersection with the $S_1 - S_2$ line then determines the highest value of $g$ where the solid is still locally stable: $g_{sh} = -6.25$. The interval $\Delta g_{sh} = g_{sh} - g_F = 0.25$ agrees well with the value 0.25 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 $S(\vec{q})$ diverges. From the data in Ref. [4] we expect that the interval $\Delta g_{sc} = g_F - g_{sc}$ is of the same size as $\Delta g_{sh}$. The supercooling and the superheating data from the same reference indicate that there is a well defined critical value of Abrikosov ratio $< \beta_A > \approx 1.18$ 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 this range becomes $\gg l$ as density modulation increases. This phase
should be distinguished from other phases described in the literature [16]. True supercon-
ducting transition in high magnetic field is almost always due to disorder, and, unless the
disorder is very strong, it will be much below the freezing transition discussed here. Finally,
the weak interlayer coupling in HTS and similar systems will act 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 easily estimated [8].

To conclude, we have presented a density-functional theory of the vortex liquid-to-solid
phase transition in homogeneous quasi 2D superconductors. As the input, we have used
the temperature-dependent first two peaks of the numerically determined structure factor
for the zeroes of the superconducting order parameter. As the output, we obtain the value
of transition temperature, the discontinuities of thermal average of the Abrikosov ratio and
of the average superfluid density, the Debye-Waller factor at the point of transition and
the latent heat. We determine the temperature interval for superheating of the vortex-solid
phase and conjectured that there is a critical point that can be reached by supercooling.
The results are found to agree well with Monte Carlo simulations. 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 $|\Psi(\vec{r})|^2$ [17] could have been used
instead so that the theory is kept entirely at the analytic level.

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Figure 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, and triangles mark the points where the solid first becomes locally stable. The line with positive slope depicts $S_1 - S_2$ relation inferred from MC simulations [13]. The transition occurs at the intersection of this line with the line which joins the crosses.
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