

Theory of Equilibrium Flux Lattices in Unconventional Superconductors

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We investigate equilibrium flux lattice structures in superconductors with unconventional order parameters, such as high- T_c cuprates, using a generalized London model with nonlocal electrodynamics derived from a simple microscopic model. We find a rich phase diagram containing triangular, centered rectangular, and square lattices with various orientations relative to the ionic lattice, as a function of magnetic field and temperature. [S0031-9007(97)03917-3]

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Existence of a mixed state, characterized by a regular array of magnetic flux lines penetrating the material, is perhaps one of the most striking properties of type II superconductors. The original pioneering work of Abrikosov [1], based on the solution of Ginzburg-Landau (GL) equations near the upper critical field H_{c2} , predicted a triangular flux lattice (FL). This prediction was subsequently verified by low field magnetic decoration experiments on a variety of conventional superconductors. In some compounds neutron scattering experiments revealed deviations from perfect triangular lattices in stronger fields [2] which were attributed to anisotropies in the electronic band structure and other effects and were modeled by GL theories containing additional higher order derivative terms reflecting the material anisotropies [3].

One would expect even richer behavior of flux lattices in the new class of heavy fermion and copper-oxide superconductors as these exhibit highly anisotropic electronic structures and, very likely, order parameters with unconventional symmetries involving nodes in the gap. In high- T_c cuprates much of the experimental and theoretical effort has been focused on the sizable region of the phase diagram just below $T_c(H)$ in which the vortex lattice properties are dominated by thermal fluctuations [4]. While understanding the physics of this fluctuation dominated regime poses an intriguing and difficult statistical mechanics problem, investigation of the equilibrium vortex lattice structures at low temperatures may provide clues about the microscopic mechanism in these materials. So far such investigations have been limited to the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compound [5,6], revealing vortex lattices with centered rectangular symmetry and various orientations relative to the ionic lattice. These have been modeled by phenomenological GL theories appropriate for anisotropic superconductors, containing additional quartic derivative terms [7] or a mixed gradient coupling to an order parameter with different symmetry [8]. These works found structures in qualitative agreement with experiment, but their inherent shortcoming is the large number of unknown phenomenological parameters and the subsequent lack of predictive power. Also, such GL theory has only

been solved for vortex lattice near H_{c2} , which is experimentally inaccessible in cuprates away from T_c . (See, however, Ref. [9] which holds promise for full solution at any field.) We have recently formulated a generalized London model [10] which is valid in an experimentally accessible region of intermediate fields $H_{c1} < H \ll H_{c2}$. This model is also phenomenological, and it contains one unknown parameter which controls the strength of the symmetry breaking term. With increasing magnetic field this model predicts a continuous transition from triangular to square FL. While no direct experimental evidence exists in cuprates at present to confirm such a prediction, a similar transition has recently been observed in a borocarbide material $\text{ErNi}_2\text{B}_2\text{C}$ [11] and has been described by a similar London model [12].

In this Letter we present a microscopic derivation of the generalized nonlocal London model for an unconventional superconductor. Based on this model we formulate, for the first time, *quantitative* and largely *parameter free* predictions for the behavior of the vortex lattice structure as a function of temperature and magnetic field. Our theory is valid in a large part of the H - T phase diagram, only restricted by the inherent domain of validity of the London model, $H \ll H_{c2}$, and T low enough that the thermal fluctuations are unimportant. The central result of this work is a prediction that the FL geometry in unconventional superconductors will display a rich and distinctive behavior as a function of field and temperature, undergoing a series of transitions and crossovers, and eventually attaining a *universal* limit at $T = 0$. The London free energy in this limit is nonanalytic, and its long wavelength part is fully determined by the nodal structure of the gap function. Such behavior is caused by the low-lying quasiparticle excitations within the nodes and thus could never occur in conventional superconductors with anisotropic band structures.

In general, the relation between the supercurrent \mathbf{j} and the vector potential \mathbf{A} of the magnetic induction $\mathbf{B} = \nabla \times \mathbf{A}$ is nonlocal in a superconductor, reflecting the finite spatial extent of the Cooper pair $\sim \xi_0$ [13]. Nonlocal corrections to physical quantities, such as the

effective penetration depth, will be of order κ^{-2} , where $\kappa \equiv \lambda_0/\xi_0$ is the GL ratio and λ_0 is the London penetration depth. For strongly type II materials ($\kappa \gg 1$) such corrections are negligible. Since cuprate superconductors fall well within this class (κ is in excess of 50 for most) local electrodynamics is always used. However, a closer examination suggests that this might not be justified in all situations, if, as it is widely believed, these materials exhibit nodes in the gap. In such a case in place of the usual coherence length $\xi_0 = v_F/\pi\Delta_0$ one is forced to define an angle dependent quality, $\xi_0(\hat{\mathbf{p}}) = v_F/\pi\Delta_{\hat{\mathbf{p}}}$, which diverges along the nodes. Clearly, in the vicinity of nodes the condition $\lambda_0/\xi_0(\hat{\mathbf{p}}) \gg 1$ is no longer satisfied and, in fact, the extreme *nonlocal* limit is achieved. Nonlocal corrections therefore cannot be dismissed in unconventional superconductors, especially at low temperatures when quasiparticles selectively populate nodal regions. From the above argument it is also clear that such corrections will be highly anisotropic and will in general break the rotational symmetry of the flow field around the vortex, contributing an anisotropic component to the intervortex interaction in the mixed state. Thus, on very general grounds, one may expect nontriangular FL structures in unconventional superconductors. We now illustrate this idea by computing the FL geometry for the simplest case of a $d_{x^2-y^2}$ superconductor with cylindrical Fermi surface in the clean limit.

The nonlocal relation between \mathbf{j} and \mathbf{A} is conveniently written in Fourier space [13]

$$\mathbf{j}_{\mathbf{k}} = -(c/4\pi)\hat{\mathbf{Q}}(\mathbf{k})\mathbf{A}_{\mathbf{k}}. \quad (1)$$

Here $\hat{\mathbf{Q}}(\mathbf{k})$ is the electromagnetic response tensor which can be computed, within the weak coupling theory, by generalizing the standard linear response treatment of Gorkov equations [14] to an anisotropic gap. We find

$$Q_{ij}(\mathbf{k}) = \frac{4\pi T}{\lambda_0^2} \sum_{n>0} \left\langle \frac{\Delta_{\hat{\mathbf{p}}}^2 \hat{v}_{Fi} \hat{v}_{Fj}}{\sqrt{\omega_n^2 + \Delta_{\hat{\mathbf{p}}}^2} (\omega_n^2 + \Delta_{\hat{\mathbf{p}}}^2 + \gamma_{\mathbf{k}}^2)} \right\rangle, \quad (2)$$

where $\gamma_{\mathbf{k}} = \mathbf{v}_F \cdot \mathbf{k}/2$, London penetration depth $\lambda_0^{-2} = 4\pi e^2 v_F^2 N(0)/c^2$, Matsubara frequencies $\omega_n = \pi T(2n - 1)$, and the angular brackets mean the Fermi surface averaging. Equation (2) is valid for arbitrary Fermi surface and gap function. For isotropic gap one recovers an expression recently derived by Kogan *et al.* from the Eilenberger theory [15]. One may simplify solving the London equation by writing it in terms of magnetic induction only. Eliminating \mathbf{j} from Eq. (1) using the Ampère's law $\mathbf{j} = (c/4\pi)\nabla \times \mathbf{B}$, one obtains

$$\mathbf{B}_{\mathbf{k}} - \mathbf{k} \times [\hat{\mathbf{Q}}^{-1}(\mathbf{k})(\mathbf{k} \times \mathbf{B}_{\mathbf{k}})] = 0. \quad (3)$$

For many purposes it is also convenient to write down the corresponding London free energy F_L , such that $\delta F_L/\delta \mathbf{B}_{\mathbf{k}} = 0$ gives the above London equation,

$$F_L = \sum_{\mathbf{k}} [\mathbf{B}_{\mathbf{k}}^2 + (\mathbf{k} \times \mathbf{B}_{\mathbf{k}})\hat{\mathbf{Q}}^{-1}(\mathbf{k})(\mathbf{k} \times \mathbf{B}_{\mathbf{k}})]/8\pi. \quad (4)$$

It is easy to see that in the local limit $Q_{ij}(k \rightarrow 0) = \delta_{ij}\lambda^{-2}$ one recovers the usual London free energy [13]. Here $\lambda \equiv \lambda(T)$ is the temperature dependent penetration depth (given below) for which it holds that $\lambda(0) = \lambda_0$.

One may study FL structure using this formalism provided the cores occupy only a small fraction of the total volume, i.e., when $H \ll H_{c2}$ and $\kappa \gg 1$. To account for the topological winding of the phase around the core [10,13] it is then necessary to insert source terms $\rho_{\mathbf{k}}$ on the right-hand side of Eq. (3). A commonly used form is [16]

$$\rho_{\mathbf{k}} = (\phi_0/\Omega)e^{-k^2\xi^2/2}, \quad (5)$$

where the prefactor ensures proper flux quantization (ϕ_0 is the flux quantum, and Ω is the area of the FL unit cell). For a given kernel $\hat{\mathbf{Q}}(\mathbf{k})$ the FL symmetry is then determined by minimizing the Gibbs free energy $G_L = F_L - H\bar{B}/4\pi$ (where $\bar{B} = \phi_0/\Omega$ is the average induction).

At long wavelengths $\hat{\mathbf{Q}}(\mathbf{k})$ can be evaluated by expanding expression (2) in powers of $\gamma_{\mathbf{k}}^2$. The zeroth order term,

$$Q_{ij}^{(0)} \equiv \delta_{ij}\lambda^{-2} = \frac{4\pi T}{\lambda_0^2} \sum_{n>0} \left\langle \frac{\Delta_{\hat{\mathbf{p}}}^2 \hat{v}_{Fi} \hat{v}_{Fj}}{(\omega_n^2 + \Delta_{\hat{\mathbf{p}}}^2)^{3/2}} \right\rangle, \quad (6)$$

is just the temperature dependent penetration depth which, at low temperatures, has the well known T -linear behavior [17]: $\lambda^{-2} \approx \lambda_0^{-2}[1 - (2\ln 2)T/\Delta_d]$ for a $d_{x^2-y^2}$ superconductor with $\Delta_{\hat{\mathbf{p}}} = \Delta_d(\hat{p}_x^2 - \hat{p}_y^2)$ and a cylindrical Fermi surface. From now on we shall focus on this simple case. The leading nonlocal term is quadratic in k ,

$$Q_{ij}^{(2)} = -\frac{4\pi T}{\lambda_0^2} \sum_{n>0} \left\langle \frac{\Delta_{\hat{\mathbf{p}}}^2 \hat{v}_{Fi} \hat{v}_{Fj}}{(\omega_n^2 + \Delta_{\hat{\mathbf{p}}}^2)^{5/2}} \gamma_{\mathbf{k}}^2 \right\rangle. \quad (7)$$

The expression $Q_{ij} = \delta_{ij}\lambda^{-2} + Q_{ij}^{(2)}$ is easily inverted to leading order in k : $Q_{ij}^{-1} \approx \lambda^2[\delta_{ij} - \lambda^2 Q_{ij}^{(2)}]$. Substituting this into Eq. (4) and specializing to fields along the z direction we have

$$F_L = \sum_{\mathbf{k}} B_{\mathbf{k}}^2 [1 + \lambda^2 k^2 + \lambda^2 \xi^2 (c_1 k^4 + c_2 k_x^2 k_y^2)]/8\pi. \quad (8)$$

Here $\xi = v_F/\pi\Delta_d$ and Δ_d is assumed to be a temperature dependent solution to the appropriate gap equation. Dimensionless coefficients c_1 and c_2 are given by

$$c_{\mu} = \frac{\lambda^2}{\lambda_0^2} \pi^3 \Delta_d^2 T \sum_{n>0} \frac{1}{2\pi} \int_0^{2\pi} d\theta \frac{\Delta_{\hat{\mathbf{p}}}^2 w_{\mu}}{(\omega_n^2 + \Delta_{\hat{\mathbf{p}}}^2)^{5/2}}, \quad (9)$$

where $w_1 = \hat{v}_{Fx}^2 \hat{v}_{Fy}^2$, $w_2 = (\hat{v}_{Fx}^2 - \hat{v}_{Fy}^2)^2 - 4\hat{v}_{Fx}^2 \hat{v}_{Fy}^2$, and the Fermi surface has been explicitly parametrized by the angle θ between $\hat{\mathbf{p}}$ and the x axis, $\hat{\mathbf{v}}_F = (\cos \theta, \sin \theta)$ and $\Delta_{\hat{\mathbf{p}}} = \Delta_d \cos 2\theta$. Coefficients c_1 and c_2 depend on temperature through a dimensionless parameter

$t \equiv T/\Delta_d$. From Eq. (9) one can deduce their leading behavior in the two limiting cases: for $t \ll 1$ we find

$$c_1 = \frac{\pi^2}{8} \frac{\lambda^2}{\lambda_0^2} \frac{1}{t}, \quad c_2 = -4c_1, \quad (10)$$

and for $t \gg 1$ (i.e., near T_c)

$$c_1 = \alpha \frac{\lambda^2}{\lambda_0^2} \frac{1}{t^4}, \quad c_2 = 8c_1, \quad (11)$$

where $\alpha = \zeta(5)(1 - 2^{-5})/8\pi^2 = 0.01272$. In the above λ also depends on t , but this will be unimportant for the following qualitative discussion.

The free energy (8) formally coincides with the one deduced previously from phenomenological considerations [10]. An interesting new feature emerging from the present microscopic model is a sign reversal of c_2 at intermediate t implied by Eqs. (11) and (10). At high temperatures c_2 is found to be positive, in agreement with [10]. It has been shown that such a term leads to centered rectangular FL structure with principal axes oriented along x or y axes of the ionic lattice [cf. Fig. 1(a)]. The magnitude of the distortion from a perfect triangular lattice ($\beta = 60^\circ$) is controlled by the magnitude of c_2 and grows with increasing magnetic field. Equation (11) shows that at fixed field this distortion will initially grow with decreasing temperature. At low temperatures Eq. (10) predicts $c_2 < 0$. This will lead to the same centered rectangular FL rotated by 45° [cf. Fig. 1(b)]. Numerical evaluation of Eq. (9) shows that c_2 passes through zero at $t^* \approx 0.19$. At this temperature the free energy (8) is isotropic and the FL will be triangular at all fields. The sign reversal of c_2 reflects the competition between the two terms of different symmetry in w_2 and is a unique consequence of the gap function having nodes.

Another consequence of nodes is the fact that, as it can be seen from Eq. (10), both c_1 and c_2 diverge as $1/t$ for $t \rightarrow 0$. This divergence signals that the response tensor $\hat{Q}(\mathbf{k})$ is a *nonanalytic* function of \mathbf{k} at $T = 0$ and the expansion in powers of $\gamma_{\mathbf{k}}^2$ breaks down. Formally this is caused by the fact that at $T = 0$ at the nodal point the expression (2) for $Q_{ij}(\mathbf{k})$ contains a term proportional to $1/\gamma_{\mathbf{k}}^2$. At $T = 0$ the frequency sum in (2) becomes an

integral which can be evaluated exactly with the result

$$Q_{ij}(\mathbf{k}) = \frac{1}{\lambda_0^2} \left\langle \hat{v}_{Fi} \hat{v}_{Fj} \frac{2 \operatorname{arcsinh} y}{y \sqrt{1+y^2}} \right\rangle, \quad (12)$$

where $y = \gamma_{\mathbf{k}}/\Delta_{\hat{\mathbf{p}}}$. For small k the dominant contribution to the angular average comes from the close vicinity of nodes and can be evaluated by linearizing $\Delta_{\hat{\mathbf{p}}}$ around the nodes. One finds that the leading nonlocal contribution is *linear* in k rather than quadratic. For $Q_{ij} = \delta_{ij} \lambda_0^{-2} + Q_{ij}^{(1)}$, we have $Q_{xx}^{(1)} = Q_{yy}^{(1)} = -\mu(k_{>} \xi_0)$ and $Q_{xy}^{(1)} = Q_{yx}^{(1)} = -\mu(k_{<} \xi_0) \operatorname{sgn}(\hat{k}_x \hat{k}_y)$, where $k_{>} = \max(|k_x|, |k_y|)$ and $k_{<} = \min(|k_x|, |k_y|)$. Prefactor $\mu = \pi^2/8\sqrt{2} = 0.8723$ is exact in the sense that all corrections to Q_{ij} are $O(k^2)$. The resulting free energy at $T = 0$ is

$$F_L = \sum_{\mathbf{k}} B_{\mathbf{k}}^2 [1 + \lambda_0^2 k^2 + \mu(\lambda_0^2 \xi_0) k_{>} (k_{>}^2 - k_{<}^2)] / 8\pi. \quad (13)$$

The nonlocal term is clearly nonanalytic in k . Its functional form is universal in the sense that it is independent of the Fermi surface structure (as long as it has tetragonal symmetry) and the prefactor μ only depends on the angular slope of the gap function and Fermi velocity at the node. Numerical evaluation shows that the free energy (13) gives rise to a centered rectangular FL structure, aligned with x or y axes, but now with the apex angle $\beta < 60^\circ$, depending *nonmonotonically* on the field. This suggests that there will be an additional transition at low temperature related to the nonanalytic behavior of the response tensor.

In order to map out the complete equilibrium H - T phase diagram we have carried out a numerical computation of the FL structure using the full expression for the response tensor $\hat{Q}(\mathbf{k})$, as given by Eqs. (2) and (12). We

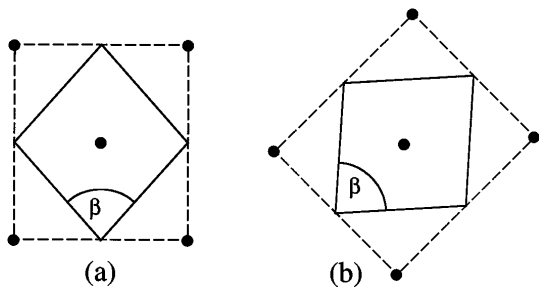


FIG. 1. Two high symmetry orientations of centered rectangular unit cell.

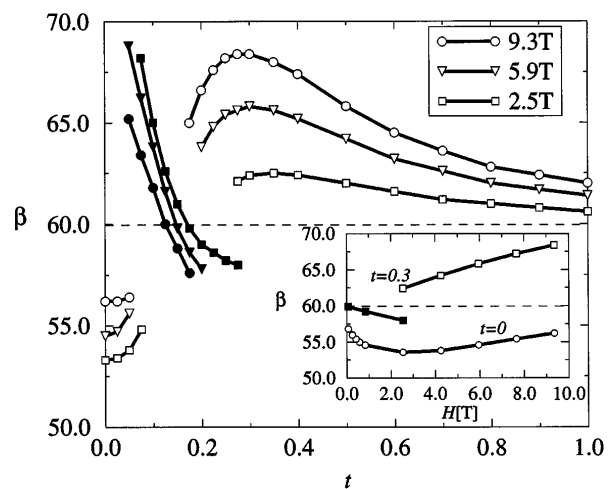


FIG. 2. Equilibrium angle β as a function of reduced temperature $t = T/\Delta_d$ for various fields. Open symbols mark lattice with orientation along x or y direction while solid symbols mark the lattice rotated by 45° . We use $\lambda_0 = 1400 \text{ \AA}$ and $\kappa = 68$. Inset: β as a function of field at fixed T .

find that free energy has two local minima for centered rectangular lattices aligned with two high symmetry directions shown in Fig. 1, as expected from the tetragonal symmetry of the problem. Which of the two becomes the global minimum depends on temperature and field. The results are summarized in Fig. 2. For high temperatures the exact result agrees well with the one obtained from the long wavelength free energy (8). The deformation of the lattice from perfect triangular grows with decreasing temperature, reaches a maximum, and then falls. Maximum distortion occurs around $t \approx 0.3$, attaining $\beta \approx 70^\circ$ at 10 T. Extrapolating this field dependence (see inset to Fig. 2), the FL should become square around $H \approx 30$ T, but this field is outside the domain of validity of the London model. At lower temperatures the distortion decreases but instead of going all the way back to triangular at t^* , the lattice undergoes a first order phase transition to another centered rectangular lattice rotated by 45° and with $\beta < 60^\circ$. Further decrease of temperature causes the angle to grow again. We note that the precise temperature at which it crosses 60° depends on field, but for all fields is close to $t^* = 0.19$, as predicted by the long wavelength approximation. At a yet lower temperature we predict another first order transition to a centered rectangular lattice along x (or y) with $\beta < 60^\circ$. The free energy difference between the two minima is very small in the region where the 45° rotated lattice wins. It is thus likely that the real system will remain in the metastable state and the experiment would detect only a smooth crossover from a lattice with $\beta > 60^\circ$ to the one with $\beta < 60^\circ$.

The present calculation can be easily generalized to treat the effects of Fermi surface anisotropy. As mentioned above tetragonal anisotropy will not modify the $T \rightarrow 0$ universal behavior but may lead to quantitative changes at higher temperatures. Orthorhombic anisotropy, on the other hand, will modify even the $T \rightarrow 0$ limit. We expect that it will, to leading order, merely rescale the coordinate axes, leading to the same structures as described above stretched by the appropriate factor [10]. It may further remove the degeneracy between two equivalent lattices related by 90° rotation. Another source of anisotropy neglected in our calculation is the nonlinear Meissner effect studied by Yip and Sauls [18], associated with the shift of quasiparticle spectrum due to the superflow. Within the quasiclassical picture this would contribute terms $\sim (|\partial_{x'} B|^3 + |\partial_{y'} B|^3)$ to the London free energy at $T = 0$, where x' and y' are 45° rotated coordinates. Our initial numerical results [19] indicate that, while not completely negligible, these terms will not significantly alter the behavior of FL reported in Fig. 2. Also neglected in our calculation is the effect of electronic disorder, which will remove the nonanalyticity of $\hat{\mathbf{Q}}(\mathbf{k})$ at longest wavelengths, just as small finite temperature would. Since the FL is most sensitive to $\hat{\mathbf{Q}}(\mathbf{k})$ at finite $k \sim l^{-1}$ (l is the vortex spacing), we expect our predictions to be robust with respect to weak disorder.

In conclusion, we have described distinctive features of the vortex lattice geometry associated with unconventional pairing and nonlocal response. Near T_c our predictions are consistent with the existing phenomenological work [7,8,10], while at low T we predict novel effects intimately related to the nodal structure of the order parameter, which are not contained in GL-type theories. These will occur simultaneously with other unique effects predicted previously, such as the $\sim \sqrt{H}$ dependence of specific heat [20]. Existing experiments probing the FL geometry in cuprates [5,6] provide a somewhat conflicting picture, and their theoretical analysis is complicated by the orthorhombic anisotropy and presence of twin boundaries. We hope that the present theory will encourage more systematic experimental work, preferably on untwinned or tetragonal materials.

After this work was completed we learned about a paper by Kosztin and Leggett [21] which discusses effects of nonlocality on the effective penetration depth in d -wave superconductors. Where overlap exists their results appear consistent with ours.

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