Charged vortices and copper nuclear quadrupole resonance in the cuprates

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The charge density induced at $T=0$ by a vortex in a type-II superconductor is studied, starting from the Lagrangian for a nonlinear Schrödinger equation. Coupled Bernoulli and Poisson equations are solved assuming that the electrostatic screening length is much shorter than the superconducting coherence length. The sign and magnitude of the charge accumulated in the vortex core agree with recent nuclear quadrupole resonance data of Kumagai, Nozaki, and Matsuda [Phys. Rev. B **63**, 144502 (2001)] on slightly overdoped $YBa_2Cu_3O_7$.

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I. INTRODUCTION

A vortex line in a type-II superconductor is associated with electrostatic fields generated by the position-dependent Bernoulli potential. One part of this potential comes from the kinetic energy of the rotating condensate. It has been predicted long ago by London,[1] and applied to the vortex line by Vijfeijken and Staas.[2] More recently, Khomskii and Freimuth[3] considered another contribution to the Bernoulli potential that is due to the position-dependent density of superconducting electrons. The local charge modulation produced by this effect has been further studied by Blatter *et al.*[4] These authors consider a BCS superconductor and calculate the charge accumulation within the vortex core under the assumption that the superconducting coherence length ξ is much larger than the Thomas-Fermi screening length λ_{TF}. Using the BCS expression for the transition temperature T_c, the accumulated charge per layer of thickness s is given by[4]

$$Q_\xi \approx \frac{2ek_F s}{\pi^3}\left(\frac{\lambda_{TF}}{\xi}\right)^2 \frac{d\ln T_c}{d\ln\mu}, \tag{1}$$

where k_F is the Fermi wave vector, the charge $e>0$, and μ is the chemical potential. The quantity $d\ln T_c/d\ln\mu$ plays an important role as it determines the sign of Q_ξ. It is proportional to the derivative of the density of states at the Fermi level, specifying the particle-hole asymmetry due to band structure. As pointed out by Khomskii and Freimuth,[3] it is energetically favorable to transfer some charge carriers from the core to the outside region, where the amplitude of the condensate wave function is larger. If the charge carriers are electrons, and the derivative of the density of states at the Fermi level is positive, expression (1) yields $Q_\xi>0$, in agreement with this prediction.

Since $Q_\xi \propto \xi^{-2}$, high-T_c superconductors may be good candidates for the detection of vortex charge owing to their short coherence length.

Recently, Kumagai, Nozaki, and Matsuda[5] studied the accumulated vortex charge in $YBa_2Cu_3O_y$ and $YBa_2Cu_4O_8$ by measuring the nuclear quadrupole frequency of $^{63}Cu(2)$, a quantity sensitive to the charge density at the copper site of the CuO_2 planes. In the vortex state obtained by applying a field $H \ll H_{c2}$, only the nuclei outside the vortex core are detected owing to the relative smallness of the core regions. In view of the overall charge neutrality an increase (decrease) of the charge density outside the core implies a decrease (increase) of the accumulated core charge.[4] Hence, by measuring the frequency difference $\Delta\nu_Q = \nu_Q(0) - \nu_Q(H)$, information can be obtained about the quantity Q_ξ.

A nonvanishing $\Delta\nu_Q$, detected by Kumagai, Nozaki, and Matsuda[5] below T_c, is a strong evidence for vortex-induced charge density in YBCO. Most interesting and puzzling is the sign of $\Delta\nu_Q$. In underdoped $YBa_2Cu_4O_8$, $\Delta\nu_Q \sim 50$ kHz is found at $T=0$, while in slightly overdoped $YBa_2Cu_3O_7$, $\Delta\nu_Q \sim -25$ kHz. Assuming that $\Delta\nu_Q$ for $^{63}Cu(2)$ increases with the number of holes in the CuO_2 planes, positive $\Delta\nu_Q$ found in the underdoped material implies a decreased hole density outside the core and gives $Q_\xi>0$. By the same token, vortices in the slightly overdoped sample should produce $Q_\xi<0$.

According to Eq. (1), we have

$$\mathrm{sgn}(Q_\xi) = \mathrm{sgn}\left(\frac{d\ln T_c}{d\mu}\right) = -\mathrm{sgn}\left(\frac{dT_c}{d\delta}\right), \tag{2}$$

where δ is the concentration of the holes. The minus sign on the right-hand side (RHS) of this equation is due to the fact that the chemical potential decreases upon doping with holes. Now, the empirical relation between T_c and δ is such that $dT_c/d\delta>0$ for underdoped samples, while $dT_c/d\delta<0$ for the overdoped ones.[6]

With these data, Eq. (2) yields a negative (positive) vortex core charge for underdoped (overdoped) samples, in disagreement with experiment. Moreover, the observed core charge is one to two orders of magnitude above the value predicted by Eq. (1).

This disagreement prompts us to consider an alternative approach to studying the charge distribution due to a vortex. In the present paper we adopt a model of repulsively interacting Bose gas. This choice is motivated mostly by the troubling sign of the observed charge. In a BCS theory this sign is coupled, via Eq. (2), to the slope of the $T_c(\delta)$ curve. This can be traced to the particle-hole symmetry-breaking terms in the effective action.[7] These terms, being proportional to the derivative of the density of states N'_μ, link the coupling between charge density and potential to the band structure. On the other hand, the effective action of a Bose condensate contains large electrostatic coupling term that is independent of band structure.[7–9] Hence, the constraint posed by Eq. (2) is avoided.

At zero temperature, the dynamics of repulsively interacting Bose gas is described by a time-dependent nonlinear Schrödinger (TDNLS) equation: the Gross-Pitaevskii equation.[10,11] The same kind of dynamics has been proposed for superconductors, in the presence of an external gauge field, by Feynman.[8] By expressing the Lagrangian as a functional of superfluid density, Feynman derives a Bernoulli equation for the electrostatic potential.

In this paper, we study a charged vortex, starting with the same Lagrangian. By combining the equation for the Bernoulli potential with the Poisson equation, we obtain a "screened" differential equation for the potential, in which the kinetic energy and the quantum pressure act as an external source. To estimate the vortex-induced charge density, we follow an approximate method similar to that of Ref. 4. First, we assume that the electrostatic screening length is much shorter than the superfluid coherence length. In this approximation, the screening term in the Poisson equation dominates and the charge density is obtained by a straightforward iteration procedure.

The paper is organized as follows. In Sec. II we review Feynman's derivation of the Bernoulli potential and obtain the Poisson equation with screening. Solution of this equation in the presence of a vortex and derivation of the induced charge density are presented in Sec. III. Section IV contains an estimate of the charge accumulated within the vortex core and a discussion of the validity of the iteration procedure. Ramifications of our results for the dynamics of superconductors and some conjectures on charged vortices in the underdoped regime are presented in Sec. V.

II. BERNOULLI POTENTIAL AND POISSON EQUATION

The Bernoulli potential for a repulsively interacting Bose fluid can be found from a variational principle starting with the Lagrangian density for the time-independent Schrödinger equation[8]

$$\mathcal{L}=-\frac{\hbar^2}{2m^*}\left|\left(\boldsymbol{\nabla}-\frac{iq}{\hbar c}\mathbf{A}\right)\Psi\right|^2-qV(\mathbf{r})|\Psi|^2-\frac{\alpha}{2}(|\Psi|^2-\rho_0)^2, \tag{3}$$

where $\Psi(\mathbf{r})$ is the condensate wave function, $\mathbf{A}$ is the vector potential, $V(\mathbf{r})$ is the electrostatic potential, and ρ_0 is the equilibrium condensate density. For Cooper pairs in a hole-doped superconductor, the charge $q=2|e|$ and $m^*=2m$. The last term on the RHS of Eq. (3) is the compressibility energy with coefficient $\alpha=(\kappa\rho_0^2)^{-1}$, κ being the compressibility of the fluid. An explicit form for this coefficient has been found by Stone[9] using Fermi surface bosonization for fermions with localized attractive interaction

$$\alpha=\frac{m^* v_F^2}{3\rho_0}. \tag{4}$$

If we set $\Psi=\rho^{1/2}\epsilon^{i\theta}$, the Lagrangian density (3) becomes

$$\mathcal{L}=\frac{-\hbar^2}{2m^*}\left[\rho\left(\boldsymbol{\nabla}\theta-\frac{q}{\hbar c}\mathbf{A}\right)^2+\boldsymbol{\nabla}\rho^{1/2}\cdot\boldsymbol{\nabla}\rho^{1/2}\right]-q\rho V(\mathbf{r})-\frac{\alpha}{2}(\rho-\rho_0)^2. \tag{5}$$

The nuclear quadrupole resonance (NQR) experiment is conducted in an external magnetic field $H\ll H_{c2}$. Hence, for r of the order of the coherence length, we have $qA/\hbar c\boldsymbol{\nabla}\theta\ll 1$.[12] The effect of the magnetic field is to introduce a cutoff on the circulating supercurrent at r of the order of the London penetration depth λ_L. Since we are dealing with the case of $\lambda_L\gg\xi$, we neglect the vector potential in what follows.

The variational principle $\delta\mathcal{L}/\delta\rho=0$ yields, with the use of Eq. (5), an equation for the Bernoulli potential

$$V(\mathbf{r})=-\frac{\hbar^2}{2m^*q}\left[(\boldsymbol{\nabla}\theta)^2-\frac{1}{2\rho}\nabla^2\rho+\frac{1}{4\rho^2}(\boldsymbol{\nabla}\rho)^2\right]-\frac{\alpha}{q}[\rho(\mathbf{r})-\rho_0]. \tag{6}$$

The compressibility term on the RHS of this equation can be related to the potential using the Poisson equation

$$\epsilon\nabla^2V(\mathbf{r})=-4\pi q[\rho(\mathbf{r})-\rho_0], \tag{7}$$

where ϵ is the dielectric constant due to bound electrons. Combining Eqs. (6) and (7), we obtain

$$(\nabla^2-\lambda_D^{-2})V(\mathbf{r})=\frac{\hbar^2}{2m^*q\lambda_D^2}\left[(\boldsymbol{\nabla}\theta)^2-\frac{1}{2\rho}\nabla^2\rho+\frac{1}{4\rho^2}(\boldsymbol{\nabla}\rho)^2\right]=F(r), \tag{8}$$

where λ_D is a Debye-like screening length given by

$$\lambda_D^2=\frac{\epsilon\alpha}{4\pi q^2}. \tag{9}$$

Substituting for α the expression (4), we see that $\lambda_D^2\simeq\epsilon\lambda_{TF}^2$.[13]

Equation (8) has the form of a Poisson equation with screening. The last equality defines the source function $F(r)$ generated by the first term of the Lagrangian (3). It is proportional to a sum of the kinetic energy and the quantum pressure.

It is interesting to compare Eq. (8) with the screened Poisson equation recently derived from a generalization of the Ginzburg-Landau (GL) theory by Koláček and Lipavský.[14] Thus far, this is a most complete treatment of the Bernoulli potential in a BCS superconductor. In a sense, it also inspired our approach to the Bose superconductor. There are similarities and differences with respect to our Eq. (8). First, the screening potential in Ref. 14 is generated by varying the internal energy with respect to the density. In our derivation this corresponds to the variation of compressibility energy. Also their kinetic-energy term is similar except for a correction due to the density dependence of m^*. Since the density takes the status of a dynamical variable,[9] this correction does not appear in the variation of the Lagrangian (3). Like the inverse compressibility α, the effective mass m^* is a function of the equilibrium density ρ_0, which is not subject to a variation.

The most important difference between the GL theory[14] and our boson theory is that the term due to density dependence of the condensation energy is not present in Eq. (8). We note that in the work of Blatter *et al.*[4] the charge distribution around the vortex is determined mainly by this term. Being proportional to the derivative of the density of states at the Fermi energy, this term contributes only if particle-hole symmetry is being broken.

In contrast, the source term $F(r)$ in our Eq. (8) is of purely kinetic origin and its sign is independent of the particle-hole asymmetry due to band structure. Its r dependence in the presence of a vortex is derived below.

III. VORTEX-INDUCED CHARGE DENSITY

For an isolated vortex line, the condensate wave function takes the form

$$\psi(\mathbf{r})=\rho_0^{1/2}f(r)\epsilon^{i\theta}=\rho(r)^{1/2}e^{i\theta}, \tag{10}$$

where (r,θ) are polar coordinates and $f(r)$ is a real function approaching 1 as $r\to\infty$. The actual *charge* density is given by

$$\delta\rho_c(r)=q[\rho(r)-\rho_0], \tag{11}$$

where $\rho(r)$ is found by solving the coupled equations (7) and (8).

To the lowest order of iteration, we start with the density $\rho^{(0)}(r)$ corresponding to the solution of Eq. (6) in the absence of electrostatic potential. We use an approximation by Fetter[15]

$$\rho^{(0)}(r)=\rho_0\frac{r^2}{r^2+\xi^2}, \tag{12}$$

where ξ is the coherence length given by

$$\xi^2=\frac{\hbar^2}{2m^*\alpha\rho_0}. \tag{13}$$

With this ansatz, the source function $F(r)$ of Eq. (8) is approximated by

$$F^{(0)}(r)=\left(\frac{\hbar^2}{2m^*\lambda_D^2 q}\right)\frac{r^2+4\xi^2}{(r^2+\xi^2)^2}. \tag{14}$$

The solution of Eq. (8) is considerably simplified in the limit of $\lambda_D/\xi\ll 1$. Then the first term on the LHS can be neglected. Invoking the approximate source function of Eq. (14), the potential $V(r)$ is given by

$$V(r)\approx-\lambda_D^2F^{(0)}(r). \tag{15}$$

Using Eqs. (7), (11), and (15), the net charge density in the presence of a vortex is

$$\delta\rho_c(r)=\frac{\epsilon\lambda_D^2}{4\pi}\nabla^2F^{(0)}(r). \tag{16}$$

Evaluating the RHS of this equation with the use of Eq. (14), we obtain the charge density as a function of r,

$$\delta\rho_c(r)=\left(\frac{\epsilon\hbar^2}{2\pi m^*q}\right)\frac{r^4+12r^2\xi^2-7\xi^4}{(r^2+\xi^2)^4}. \tag{17}$$

This charge density satisfies the condition of perfect screening: Explicit integration of Eq. (17) over infinite volume yields exactly zero. The density is *negative* for $r<0.75\xi$ and the charge accumulated in this region is canceled by the *positive* charge from the region of $r>0.75\xi$. For $r\gg\xi$, the function (17) behaves asymptotically as r^{-4}. The latter property is also shared by the induced charge density calculated in Ref. 4.

IV. VORTEX CORE CHARGE

To obtain the charge accumulated within the core (per unit length of the vortex line), we integrate the charge density (17) over the volume of a cylinder of radius ξ,

$$Q_c=2\pi\int_0^\xi\delta\rho_c(r)r\,dr=-\frac{\epsilon\hbar^2}{2m^*q\xi^2}. \tag{18}$$

Since $q>0$, the RHS of Eq. (18) is negative. This agrees with the charge sign determined via NQR on slightly overdoped $YBa_2Cu_3O_7$.[5] Note that distinct from Eq. (1), Eq. (18) does not involve the quantity $d\ln T_c/d\ln\mu$. Nevertheless, at least for a parabolic band, Eq. (1) can be brought to a form similar in structure to Eq. (18). This is surprising in view of the fact that the charge of Eq. (1) is generated by position-dependent condensation energy whereas that of Eq. (18) comes from a combination of kinetic energy and quantum pressure.

To clarify this point, we consider in Eq. (1) the product $k_F\lambda_{\mathrm{TF}}^2$. For a free-electron gas we have[16]

$$k_F\lambda_{\mathrm{TF}}^2=\frac{4\hbar^2}{\pi me^2}. \tag{19}$$

Introducing this result into Eq. (1), we have

$$Q_\xi\simeq\frac{\hbar^2es}{2\pi^2me^2\xi^2}\,\frac{d\ln T_c}{d\ln\mu}. \tag{20}$$

On the other hand, letting $m^*=2m$ and $q=2e$, the charge per sheet according to Eq. (18) is given by

$$Q_\xi=sQ_c\simeq-\frac{\hbar^2es}{8me^2\xi^2}\,\epsilon. \tag{21}$$

For a parabolic band, we obtain with the use of the BCS expression for T_c,

$$\frac{d\ln T_c}{d\ln\mu}\cong(2gN_\mu)^{-1}, \tag{22}$$

where g is the constant of attractive interaction. We note that the quantity on the RHS of this equation is of order one. Now we see that the only distinction between Eqs. (20) and

(21) is the presence of ϵ in Eq. (21). However, if the quantity λ_{TF}^2 in Eq. (1) is replaced by $\lambda_D^2 \simeq \epsilon \lambda_{TF}^2$, even this distinction is erased.

It should be stressed that such enhancement of the screening length by polarization of bound electrons should be included in any theory of charged vortices. It becomes especially important in superconducting cuprates.[13] Since $\epsilon \sim 10-10^2$, the disagreement between the BCS theory[4] and the experimental magnitude of the core charge is removed. However, the difficulty with the charge sign remains unresolved within the BCS approach.

For a rough estimate of the magnitude of the accumulated core charge in slightly overdoped $YBa_2Cu_3O_7$ we use $\xi = 1.6 \times 10^{-7}$ cm and $s \simeq 2 \times 10^{-7}$ cm. With these values, the accumulated core charge per sheet according to Eq. (21) is

$$Q_\xi \sim -5\epsilon \times 10^{-3} e. \tag{23}$$

The core charge per sheet observed in this sample ranges from $-5 \times 10^{-3} e$ to $-2 \times 10^{-2} e$.[5] We see that Eq. (23) can explain this result with ϵ ranging from 1 to 4.

It remains to enquire about the validity of the iterative approach used to solve the Poisson equation with screening. We now show that there is a limiting value of the ratio λ_D^2/ξ^2 above which the iteration scheme breaks down. To determine this value, we investigate the actual boson density $\rho(r) = |\Psi(r)|^2$ in the presence of the charged vortex. From Eq. (12) we see that, for a neutral superfluid, this quantity is equal to zero for $r=0$. The effect of the potential in the charged superfluid is to fill the vortex core with bosons. It turns out that the iterative approach is most accurate when the core is nearly completely filled. This corresponds to the *strong* screening limit. We can see this by calculating $\rho(r=0)$ using Eqs. (11) and (17),

$$\rho(0) = \rho_0 \left(1 - \frac{7\hbar^2 \epsilon}{2\pi \rho_0 m^* q^2 \xi^4}\right) \simeq \rho_0 \left(1 - 28 \frac{\lambda_D^2}{\xi^2}\right). \tag{24}$$

We used the screening and the coherence lengths given in Eqs. (9) and (13) to obtain the expression on the RHS of Eq. (24). According to this expression the quantity $\rho(0)$ becomes negative when $\lambda_D^2/\xi^2 > \frac{1}{28}$. This signals a breakdown of the iteration scheme, since the boson density, being equal to $|\Psi|^2$, is necessarily positive or zero.

For values of $\lambda_D^2/\xi^2 > \frac{1}{28}$, the full Eq. (8) including the $\nabla^2 V(r)$ term must be solved. In the limit of $\lambda_D/\xi \gg 1$, the second term on the LHS of this equation can be neglected and the induced charge density $\delta\rho_c(r) \sim -\epsilon F^{(0)}(r)/(4\pi)$. According to Eq. (14), this expression is negative for all values of *r*. This may be of relevance for the interpretation of the NQR data on underdoped YBCO (see Sec. V).

Let us estimate the RHS of Eq. (24) for slightly overdoped $YBa_2Cu_3O_7$. From Eqs. (9) and (13) we obtain

$$\frac{\lambda_D^2}{\xi^2} \approx \frac{\epsilon \hbar^2}{64\pi e^2 \rho_0 m \xi^4}. \tag{25}$$

Using $\xi \approx 1.6 \times 10^{-7}$ cm and $\rho \approx 5 \times 10^{20}$ cm^{-3}, we obtain from Eq. (25), $\lambda_D^2/\xi^2 \approx 8 \times 10^{-5} \epsilon$. For $\epsilon \approx 10^2$, this ratio amounts to about $\frac{1}{5}$ of the critical value 3.5×10^{-2}. Since $\epsilon < 4$ is required to explain the measured core charge from Eq. (23), we see that the criterion for validity of the iteration scheme is met in this material.

V. DISCUSSION

In the preceding section, we came to the conclusion that the NQR experiment on charged vortices in slightly overdoped YBCO can be explained using a model of repulsively interacting Bose superfluid. We now discuss relevance of this result for superconducting dynamics of cuprates.

In BCS theory, there is a coupling of the hole density to the superconducting energy gap when the particle-hole symmetry is broken owing to a nonzero value of the quantity N'_μ.[7] In contrast, the Lagrangian of charged Bose superfluid contains a large electrostatic coupling term independent of N'_μ. Consequently, the sign of the charge induced in the vortex core becomes decoupled from the BCS constraint of Eq. (2). In this way, the present theory yields a core charge that is in agreement with the NQR data on $YBa_2Cu_3O_7$.[5]

If we consider a generalization of Eq. (3) to a time-dependent condensate, then the electrostatic coupling term appears in a gauge-invariant combination $\rho(qV - \partial\theta/\partial t)$. This topological term is present, since the bosonic field itself can be regarded as the order parameter.[7,9] The corresponding dynamics is that of a Galilean-invariant TDNLS equation.[8,9] Sá de Melo, Randeira, and Engelbrecht[17] studied the crossover between the weak-coupling BCS and the Bose condensation using functional integral formulation. In the extreme strong-coupling limit, they find that the dynamics is described by a Galilean-invariant TDNLS equation. However, Stone[9] has shown that the dynamics of a BCS condensate at $T=0$ is described by this dynamics even in the weak-coupling case. This result is of importance for vortex dynamics in connection with the problem of the Magnus force.[18] The agreement of the vortex charge derived from Eq. (3) with the NQR experiment on slightly overdoped YBCO lends support to the conclusions of Stone.[9] Apparently, the electrostatic potential in the gauge-invariant topological term is at work in overdoped YBCO.

It has been pointed out by Uemura[19] that overdoped cuprates are characterized by a BCS condensation corresponding to simultaneous pair formation and condensation at T_c. That Bose condensation is relevant for both underdoped and overdoped cuprates has been argued by Schneider and Pedersen.[20] These authors used the model of an interacting charged Bose gas and found compatibility with empirical trends of various thermodynamic properties of cuprate superconductors.

This brings us to the question of the sign of vortex charge observed by Kumagai, Nozaki, and Matsuda[5] in underdoped YBCO. Due to close proximity to the Mott insulator, physical properties in this regime are highly anomalous. One outstanding feature is the existence of the pseudogap regime observed below some temperature above T_c. As pointed out by Lee and Wen,[21] the pseudogap is expected to persist in the core region of a superconducting vortex. It is possible that the charge-sign anomaly is caused by this modification of the vortex core. Strong correlation physics must be used, such as the t-J model, to describe the vortex structure in the underdoped regime.[21] Another possibility is to assume that the ratio $\lambda_D/\xi \gg 1$, owing to a strong reduction of the compressibility of the Fermi gas by Coulomb correlations.[22] As shown below Eq. (24), under this assumption the charge density remains negative even far outside the vortex core. Noting that the nuclei contributing to the NQR signal come just from this region, the positive sign of $\Delta\nu_Q$ observed in the underdoped YBCO would be explained. Unfortunately, the compressibility of the cuprates near the Mott transition is still an unsettled topic, so that this idea lacks a firm numerical evidence.

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