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Pippard-coherence-length tensor for anisotropic superconductors

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In the framework of a generalized fully anisotropic Bardeen-Cooper-Schrieffer-type quantum theory, the low-temperature, low-field coherence-length tensor is derived from first principles. Anisotropy of the Fermi surface, the gap parameter, and the energy band structure in the effective-mass approximation is included. For lattice parameters and effective-mass ratios suitable for the high-T_c superconductors the coherence lengths along the principal axes show a marked anisotropy exceeding 20:1. The coherence length along the c axis is found to be of the order of the lattice spacing c, suggesting marginal two dimensionality.

I. INTRODUCTION

Anisotropic superconductivity is generally due to the anisotropy of the Fermi surface, of the coupling between the electrons (or holes) and the bosons that mediate the Cooper-pair formation, and of the boson density of states.[1] Until the recent discovery of the high-T_c superconductors,[2,3] and except for the $A15$ compounds and the heavy-fermion and organic superconductors, anisotropic effects in superconductivity had been less important, since most other superconductors are metals with cubic or hexagonal close-packed crystal symmetry and an approximately spherical Fermi surface.[4] While the fundamental work by Bardeen, Cooper, and Schrieffer (BCS theory)[5,6] included anisotropic effects at the outset, they arrived at their final results (BCS model)[5] by invoking isotropy on the just mentioned grounds. This isotropic BCS model describes properly many thermodynamic and electrodynamic properties of the now classical superconductors in the low-magnetic-field limit. The numerous subsequent anisotropic extensions were therefore restricted to the nearly isotropic case.[1,7,8]

The new high-T_c superconductors, however, possess strongly anisotropic features; the Fermi surface is, at least within the first Brillouin zone, open along the c axis.[9–11] And while at present several models have been suggested for the mediating mechanism, e.g., via phonons,[12,13] excitons,[14–17] plasmons,[18,19] or spin configurations,[20,21] the coupling potential was shown or assumed to be effective mainly in the basal plane. Thirdly, the electronic properties, specifically the effective-mass ratio determined from conductivity measurements[22] or band-structure calculations,[23] show an order-of-magnitude difference whether taken along the c axis or in the basal plane. Last, recent experiments of critical magnetic fields and critical currents display a similarly large anisotropy.[22,24] Nonetheless, most of the thermodynamic measurements on these anisotropic compounds seem to agree reasonably well[25] with predictions from the isotropic BCS model, notably, the interrelation between the critical temperature and the gap parameter $2\Delta/kT_c \sim 3.5$–4.7. But for the vectorial electromagnetic properties interconnected by tensors we should expect discrepancies. Both for measurements and their interpretation, as well as for the many potential applications of these materials, the electromagnetic properties are of paramount importance, and anisotropy could therefore provide a crucial clue to the better understanding of the high-T_c superconductors.

II. BCS COHERENCE LENGTH

It was the Meissner-Ochsenfeld effect, whereby a sample expels the embedded magnetic flux when cooled below the transition temperature, that led to the first useful phenomenological model for the electromagnetic properties. London and London[26] conjectured that there exists a linear-response law between the supercurrent $\mathbf{j}$ and the applied vector field $\mathbf{A}$, $\mathbf{j}(r) = (c_L\Lambda)^{-1}\mathbf{A}(r)$, where c_L denotes the speed of light. The London coefficient Λ is given in terms of the electronic charge e and mass m, and the charge-carrier number density n: $\Lambda = m/ne^2$. On a purely empirical basis Pippard[27] was led to a linear but nonlocal law. BCS could show that their pairing approximation indeed results in a nonlocal response for $T \to 0$ (Ref. 5)

$$\mathbf{j}(\mathbf{r}) = -\text{const} \times \int d^3\mathbf{r}' J(R,T)\mathbf{R}[\mathbf{R}\cdot\mathbf{A}(\mathbf{r}')]/R^4 , \qquad (1)$$

where $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, $R = |\mathbf{R}|$. The (scalar) spatial weight

function $J(R,T)$ falls off over a characteristic distance $\xi_0 = \int_0^\infty dR\, J(R, T=0) = \hbar^2 k_F/(\pi m \Delta_0)$, where k_F is the Fermi wave number. Pippard and Faber[27] had introduced a similar length. This correlation length that stems from microscopic electron-scattering effects is one of the fundamental scale lengths characterizing all superconductors for low temperatures in the absence of strong magnetic fields. In the vicinity of the critical temperature or the critical magnetic field, both this long-range correlation ξ, and the gap parameter Δ become very small, allowing for perturbation expansions in these parameters. This is the basis for the Ginzburg-Landau-Abrikosov-Gor'kov theory (GLAG)[28] and its numerous anisotropic extensions.[29] In addition to the very limited regime $T \to T_c$, $H \to H_c$, the validity of the GLAG theory may be extended for the dirty limit,[30] where the presence of a substantial concentration of foreign scattering centers can also reduce the coherence length.

III. ANISOTROPIC GENERALIZATION

The fundamental nature of the coherence length as a scale length makes desirable its derivation from first principles *irrespective of structural and chemical details,* but tractable for arbitrary anisotropy. The coherence length evaluated for $T \to 0$ and $H \to 0$, for the pure limit can be considered as an upper bound to ξ. We will therefore not concern ourselves with the GLAG limit, which has been treated to some extent elsewhere.[31] The derivation[5] of the function $J(R,T)$ that leads to a scalar value of ξ_0 cannot be practicably generalized to the fully anisotropic case. More generally[32,33] we will need to consider the Fourier-transformed (FT) quantities of the current and the field, since (i) the essential electronic correlations, in particular their anisotropic modifications, come about in wave number space, where (ii) the scattering processes between the carriers and the electromagnetic (EM) fields, i.e., the photons, can easily be formulated; (iii) the anisotropic penetration of EM fields into, their propagation within, and their reflection off a superconductor are governed by a dispersion relation[32,33] that naturally involves only the Fourier transform in wave-number and frequency space; (iv) the anisotropic convolution similar to Eq. (1) that is nonlocal in configuration space factorizes in wave-number space. Specifically, invoking the general properties of an FT, its limit for a small but finite photon momentum vector yields the long-range coherence tensor in configuration space.

To arrive at tractable results within an anisotropic BCS theory, we now specialize to an anisotropic effective-mass description. In the effective-mass approximation, the Hamilton operator for an electron in a periodical crystal potential can be replaced in the following manner: $\mathcal{H} = \not{p}^2/2m + V(\mathbf{x}) \to \mathcal{H}_{\text{eff}} = \frac{1}{2}\not{p}\cdot\vec{\mu}\cdot\not{p}$, where $\not{p}$ is the momentum vector operator, and $\vec{\mu}$ is the (symmetric) inverse mass tensor. Correspondingly, the (charge) current operator must then also be modified: $\not{j} = (e/2m)\not{p} + \text{H.c.} \to \not{j}_{\text{eff}} = (e/2)\vec{\mu}\cdot\not{p} + \text{H.c.}$; in other words, in the effective-mass approximation, the anisotropic crystal properties are carried out by the operators, while the eigenfunctions remain plane waves. Our choice of $\not{j}_{\text{eff}}$ is necessary (but not sufficient) for the norm of $\mathcal{H}_{\text{eff}}$ to be preserved in time.[34] Finally, to accommodate electromagnetic fields, the canonical momentum operator $\not{p}$ has to be replaced by $\not{p} - (e/c_L)\mathbf{A}$. We may note in passing, that a more general approach is possible, which, however, by the substitution $\vec{\mu}\cdot\not{p} \to \hbar^{-1}\nabla_{\mathbf{k}}\varepsilon$ would have to rely on the explicit knowledge of the energy-band dispersion throughout the Brillouin zone.[33]

By first-order perturbation theory, neglecting terms of order A^2, the expectation value of the current operator can be divided into a diamagnetic and a paramagnetic contribution, similar to BCS,

$$\langle \not{j}_{\text{eff}} \rangle = \mathbf{j}_D(\mathbf{q}) + \mathbf{j}_P(\mathbf{q}) , \tag{2}$$

where

$$\mathbf{j}_D(\mathbf{q}) = -(ne^2/c_L)\vec{\mu}\cdot\mathbf{A}(\mathbf{q}) ,$$

and

$$\mathbf{j}_P(\mathbf{q}) = [2e^2\hbar^2/(\Omega c_L)\vec{\mu}]\cdot\sum_{\mathbf{k}}(\mathbf{k}-\mathbf{q}/2)(\mathbf{k}+\mathbf{q}/2) \cdot L(\mathbf{k},\mathbf{k}-\mathbf{q})\vec{\mu}\cdot\mathbf{A}(\mathbf{q}) .$$

Here, $\mathbf{q}$ is the photon wave vector, $\mathbf{k}$ the electron wave vector, Ω the crystal volume, and $L(\mathbf{k},\mathbf{k}') \equiv L(\varepsilon_{\mathbf{k}},\varepsilon_{\mathbf{k}'})$ denotes the electron-photon scattering matrix elements.[5,35] To arrive at Eq. (2), no simplification could be made by a specific choice of gauge, as was possible for the isotropic case.[5] We may symmetrize the BCS expression for L and generalize it to a $\mathbf{k}$-dependent gap parameter Δ by the substitution $\varepsilon_0^2 \to (\Delta_{\mathbf{k}}\Delta_{\mathbf{k}'})$.[6] By introducing such a gap parameter, we have, in effect, taken into account an anisotropic pair potential, mediated by an anisotropic boson interaction, and an anisotropic boson density of states. Consequently, we may rewrite the current in the form $\mathbf{j}(\mathbf{q}) = -(c_L/4\pi)\vec{\mathbf{K}}(\mathbf{q})\cdot\mathbf{A}(\mathbf{q})$, where the paramagnetic component of the response tensor $\vec{\mathbf{K}}$ has the form

$$\vec{\mathbf{K}}_P(\mathbf{q}) = -[8\pi e^2\hbar^2/(c_L^2\Omega)]\vec{\mu}\cdot\sum_{\mathbf{k}}\mathbf{k}\mathbf{k}\cdot L(\mathbf{k}+\mathbf{q}/2,\mathbf{k}-\mathbf{q}/2)\vec{\mu}.$$

To arrive at this, we made use of the reversal symmetry of $L(\mathbf{k},\mathbf{k}') = L(\mathbf{k}',\mathbf{k})$, and further imposed inversion symmetry that is apparently common to all superconducting materials,[4] $L(\mathbf{k},\mathbf{k}') = L(\pm\mathbf{k},\pm\mathbf{k}')$. The vectorial FT of the current then yields an expression similar to Eq. (1). The long-range properties in configuration space of this expression determine the coherence-length tensor. But the long-range behavior in configuration space is also given by the short-range properties of the kernel $\vec{\mathbf{K}}(\mathbf{q})$, i.e., by its limit for small but finite $\mathbf{q}$. Expanding, therefore, the matrix element to second order in $\mathbf{q}$ yields

$$L(\mathbf{k}+\mathbf{q}/2,\mathbf{k}-\mathbf{q}/2) \to -\partial_E f + \tfrac{1}{2}(\varepsilon_1\Delta - \Delta_1\varepsilon)^2(1-2f)E^{-5} .$$

Here, the first term $-\partial_E f$ is the familiar result of BCS that vanishes for $T \to 0$, while the electron-energy dispersion $\varepsilon_1 = \frac{1}{2}\mathbf{q}\cdot\nabla_{\mathbf{k}}\varepsilon$, and similarly the gap dispersion Δ_1, enter the second term. The matrix element is still fully $\mathbf{k}$ dependent via ε_1, Δ_1, Δ, the electron energy (with respect to the Fermi level) ε, and the quasiparticle energy, $E = +(\varepsilon^2+\Delta^2)^{1/2}$, and temperature dependent via the

Fermi distribution function $f(E)$. This term remains finite as $T \rightarrow 0$. We have suppressed additional terms of order $q^2 \partial_E^n f(E)$, containing first and higher order derivatives of f.[33] This is legitimate for the low-temperature regime. Regrouping $\vec{\vec{K}}$ into a $\mathbf{q}$-independent and, via ε_1 and Δ_1, a $\mathbf{q}$-dependent term results in

$$\vec{\vec{K}} = \frac{4\pi n e^2}{c_L^2} \vec{\mu} \cdot \left[\left(\vec{\vec{I}} + \frac{2\hbar^2}{n\Omega} \sum_{\mathbf{k}} \mathbf{k}\mathbf{k} \cdot \partial_E f \vec{\mu} \right) - \frac{\hbar^2}{n\Omega} \sum_{\mathbf{k}} \mathbf{k}\mathbf{k} \cdot (\Delta\varepsilon_1 - \varepsilon\Delta_1)^2 \frac{1-2f}{E^5} \vec{\mu} \right].$$

As usual, $\vec{\vec{I}}$ is the unit tensor. We can identify the first term as the tensor $\vec{\vec{\Lambda}}_T^{-1}(T)$, corresponding to the BCS (scalar) temperature-dependent London coefficient, whose nonzero limiting value for $T \rightarrow 0$ accounts for the existence of the Meissner-Ochsenfeld effect.[5] This tensor $\vec{\vec{\Lambda}}^{-1}$ determines the anisotropic penetration depth in the London Limit ($\mathbf{q} \rightarrow \mathbf{0}$) in terms of the anisotropic Drude plasma frequency.[33]

IV. COHERENCE-LENGTH TENSOR

The second term, on the other hand, gives rise to *long-range electronic correlations.* We rewrite it in Cartesian coordinates in terms of the fourth-rank correlation-length tensor as $q_i q_j \Xi_{ijnn'} = q_i q_j \xi_i \xi_j$, with

$$\Xi_{ijnn'} = \frac{\hbar^2}{4n\Omega} \sum_{m=1}^{3} \mu_{mn'} \sum_{\mathbf{k}} k_n k_m (\Delta \partial_{k_i} \varepsilon - \varepsilon \partial_{k_i} \Delta) \times (\Delta \partial_{k_j} \varepsilon - \varepsilon \partial_{k_j} \Delta) \frac{1-2f}{E^5}. \quad (3)$$

The indices n and n' denote the vector components of $\mathbf{j}$ and $\mathbf{A}$, respectively. For the isotropic BCS case, which moreover assumes $\partial_{\mathbf{k}} \Delta \equiv 0$, i.e., $\Delta \rightarrow \Delta_0 = \text{const}$, we have at once, at absolute zero, $\Xi \rightarrow (\hbar^2 k_F / 2m\Delta_0)^2 \sim \xi_0^2$ for purely transverse waves, i.e., $\mathbf{q} \cdot \mathbf{A} \equiv 0$. Before evaluating the principal correlation tensor components for tetragonal or nearly tetragonal crystal structure[36-38] and the square-shape open Fermi surface,[9-11] we discuss the various contributions to Ξ. The thermal weight function $(1-2f)$ is identical to the one found in the integral equation that determines the gap parameter[39] from the interaction potential $V_{\mathbf{k},\mathbf{k}'}$. We therefore expect that, generally, the overall magnitude of Ξ diminishes with increasing T similar to Δ. If the interaction potential $V_{\mathbf{k},\mathbf{k}'}$ is effective mainly in the basal plane, the resulting Δ will depend only on $\mathbf{k}_\perp$, as long as the Fermi surface curvature along k_z is small. The reason for this is that we can estimate the corrections in k_z of the quasiparticle energy to be at most of the order of $(m_\perp/m_z)(k_{z\mathrm{BZ}}/k_{\perp F})^2 \sim 10^{-3}$, using a mass ratio characteristic for the high-T_c superconductors, $m_z/m_\perp \sim 5$-10,[22-24] a wave vector at the Brillouin-zone boundary $k_{z\mathrm{BZ}} = \pi/c$, and an average Fermi wave vector in the basal plane $k_{\perp F} \sim \pi/a$, with $a/c \sim 0.3$.[36] (This argument assumes that umklapp processes are unimportant.[40]) If, moreover, the potential V does not vary too drastically with $\mathbf{k}_\perp$, a basal-plane–averaged V will lead to essentially the same Δ-T_c relation as in the isotropic case of BCS. The dispersion of Δ with $\mathbf{k}$ enters the bracket terms in Eq. (3) that determine the overall strength of the correlation. The BCS-like term $\Delta \nabla_{\mathbf{k}} \varepsilon = \Delta \hbar \mathbf{v}$ may be reduced or augmented by the gap dispersion via the new term, $\varepsilon \nabla_{\mathbf{k}} \Delta$. However, this term is small right at the Fermi surface, where $\varepsilon \rightarrow 0$. If we can assume a small gap dispersion, only the immediate vicinity of the Fermi surface will make a substantial contribution to the $\mathbf{k}$-space sum in Eq. (3) because of the strongly weighting cofactor E^{-5}.

We now evaluate the principal correlation lengths at zero temperature. Along the [001] direction, i.e., for $q_z \neq 0$, we get for transverse modes,[32] invoking the mean-value theorem that holds definitely for a small-gap dispersion

$$\Xi_{zz,xx} + \Xi_{zz,yy} \sim \frac{\hbar^6 \Delta_0^2 k_{\perp F}^2}{8nm_\perp m_z^2} \frac{1}{\Omega} \sum_{k_z} k_z^2 \sum_{\mathbf{k}_\perp} E^{-5}(\mathbf{k}_\perp) \sim \xi_z^2.$$

Thus, $\xi_z \sim (1/3\sqrt{2}) \hbar^2 k_{z\mathrm{BZ}} / (m_z \Delta_0)$. For the correlation length along the [100] axis we have similarly

$$\xi_x = \frac{\pi}{8\sqrt{3}} \frac{\hbar^2 k_{\perp F}}{m_\perp \Delta_0} \left(1 + \frac{32}{3\pi^2} \frac{m_\perp}{m_z} \frac{k_{z\mathrm{BZ}}^2}{k_{\perp F}^2} \right)^{1/2} \sim \frac{\pi}{8\sqrt{3}} \frac{\hbar^2 k_{\perp F}}{m_\perp \Delta_0}.$$

Thus, we obtain $\xi_\perp / \xi_z \sim (k_{\perp F}/k_{z\mathrm{BZ}})(m_z/m_\perp) \sim (c/a) \times (m_z/m_\perp)$, which is of order 20 or greater.

V. DISCUSSION

For an effective-mass ratio $m_z/m_\perp \sim 10$ (Refs. 22-24) and a critical temperature of 50 K (100 K), the coherence length along the c axis is of the order of 20 Å (10 Å), i.e., of the same order as the lattice parameter c.[36-38] This very short correlation length along [001] is essentially due to the large effective-mass ratio $m_z/m_\perp$ and the small aspect ratio $k_{z\mathrm{BZ}}/k_{\perp F}$ of the effective Fermi surface within the first Brillouin zone, neglecting umklapp processes along the c axis. The small value of ξ_z suggests that the superconductivity in the high-T_c superconductor is only marginally three dimensional.

The Pippard coherence length ξ characterizes the spatial extent of the electronic correlations in a superconductor. It is to be compared with other pertinent lengths, such as the sample dimension d (e.g., the film thickness), the depth δ of the insulating layer in a tunneling junction, the electronic mean free path l, or the (London) penetration length λ for static electric and magnetic fields. Usually, one has $d \gg \xi$, $\delta \ll \xi$, while the superconductor is called clean, or dirty, for $l \gg \xi$, or $l \sim \xi$, respectively. Superconductors of type I (type II) have a small (large) Ginzburg-Landau parameter, $\kappa = \lambda/\xi < 2^{-1/2}$ ($> 2^{-1/2}$) in the isotropic case. Our results suggest, therefore, that superconductivity along the c axis will be definitely of type II, even at absolute zero. In any case, the use of any one single, isotropic coherence length becomes rather doubtful for such large anisotropy ratios.

It is difficult to determine the coherence length directly from experiment. Rather, it is usually inferred by relying on certain model assumptions. In the framework of GLAG, the fluctuations near the critical transition and

the critical fields emerge as functions of ξ. Based on this, for the new superconductors coherence lengths were recently determined.[41,42] From the specific-heat fluctuation measurements[42] only an isotropic ξ was derived since the invoked model contained only a scalar coherence length. The values quoted range from 7 Å for ξ_z to 34 Å for $\xi_\perp$, in excellent agreement with our value for ξ_z, while the experimental value for $\xi_\perp$ appears to be smaller by about a factor of two to four, compared to our results. Moreover, our scaling for the ratio of the Ginzburg-Landau parameters, $\kappa_z/\kappa_\perp = (m_z/m_\perp)^{3/4}(k_{zBZ}/k_{\perp F})^{1/2}$, differs somewhat from the one quoted in Ref. 41 in its additional dependence on the configuration of the Fermi surface. For the given parameters, although, this would result in no appreciable numerical discrepancy. The smaller experimentally determined value for $\xi_\perp$ is probably due to a very short mean free path that then would be dominant as long as $\xi_\perp > l$. This is probably the case for these high-T_c ceramics, even for single crystals.

An interesting aspect may arise further in tunneling as a result of the very small ξ_z encountered here, when the insulating layer is applied in the *a-b* plane. Traditionally, pair tunneling can only take place when the barrier thickness is much smaller than ξ. For the small $\xi_z \sim 10$ Å given above, this is most likely to be violated. On the other hand, layer-to-layer tunneling *without* a barrier, and other correlation effects along the *c* axis, are conceivable, since ξ_z is equal to or less than the lattice-translation vector c. The recently reported point-contact tunneling experiments on thin films show striking equidistant multiple-gap features[43,44] with exponentially decaying strength, possibly connected to such correlations.

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