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Proposal for an experiment to test a theory of high-temperature superconductors

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A theory for the phenomena observed in copper-oxide based high-temperature superconducting materials derives an elusive time-reversal and rotational symmetry-breaking order parameter for the observed pseudogap phase ending at a quantum-critical point near the composition for the highest T_c. An experiment is proposed to observe such a symmetry breaking. It is shown that angle-resolved photoemission yields a current density which is different for left and right circularly polarized photons. The magnitude of the effect and its momentum dependence is estimated. Barring the presence of domains of the predicted phase, an asymmetry of about 0.1 is predicted at low temperatures in moderately underdoped samples.

I. INTRODUCTION

Despite twelve years of intensive experimental and theoretical studies of copper-oxide based superconducting compounds,[1] no consensus about the fundamental physics or even about the minimum necessary Hamiltonian to describe the phenomena has emerged. One of the few theoretical ideas which has clearly survived experimental tests is that at density $x \approx x_c$ near that for the maximum superconducting transition temperature, the normal state is a marginal Fermi liquid (MFL).[2] The MFL is characterized by a scale-invariant particle-hole fluctuation spectrum which is only very weakly momentum dependent. One of the predictions of MFL hypothesis is that the single-particle spectral function $G(\mathbf{k},\omega)$ has a nearly momentum-independent self-energy proportional to $\max(|\omega|,T)$. The frequency and temperature dependence as well as the momentum independence have been tested in angle-resolved photoemission experiments.[3]

The observed non-Fermi-liquid behavior near $x \approx x_c$ in resistivity, thermal conductivity, optical conductivity, Raman scattering, tunneling spectra, and the Cu nuclear relaxation rate follow from the MFL hypothesis. The scale invariance of the MFL fluctuations implies that a quantum critical point (QCP) exists at $x = x_c$, near the optimum composition. One expects that, in two or three dimensions, the QCP at $T=0$ is the end-point of a phase of reduced symmetry as x is varied. Similarly a line of transitions or at least a crossover is expected at a finite temperature $T_p(x)$ terminating at $(x = x_c, T=0)$. Indeed the generic phase diagram, Fig. 1, of the copper-oxide compounds around $x \approx x_c$ displays such a topology. Region I has MFL properties dominated by quantum fluctuations, region III displays properties characteristic of a Fermi liquid, while region III—the pseudogap region—displays a loss of low-energy excitations compared to region II. Below the line $T_p(x)$ between regions I and II, the single-particle spectrum displays lowered rotational symmetry, while no translational symmetry appears broken. The superconductivity region sits spanning the three distinct normal-state regions.

Figure 1 may be compared to the topologically similar phase diagram of some heavy-fermion compounds, in which the line $T_p(x)$ corresponds to an antiferromagnetic transition.[4] From this point of view the crucial question in Cu-O compounds is the symmetery in region II of the so-called pseudogap phase.

A systematic theory[5,6] starting with a general model for Cu-O compounds provides an answer to this question. Region II in Fig. 1 is derived to be a phase in which a fourfold pattern of current flows in the ground state in each unit cell as shown in Fig. 2. Time-reversal symmetry as well as rotational symmetry is broken but the product of the two is conserved. This phase has been called the circulating-current (CC) phase. Quantum fluctuations about this phase are shown to have MFL fluctuations, characteristic of region I. The same fluctuations promote ''*d*'' or generalized ''*s*''-state pairing depending on the Fermi surface at a given doping.

While a microscopic theory in agreement with most of the principal experimental results has been presented, one can be confident of the applicability of the theory only if the CC phase is directly observed. The CC phase has a very elusive-order parameter. The fourfold pattern of microscopic magnetic moments in each unit cell changes the Bragg intensity for polarized neutrons at certain preexisting Bragg spots. But the intensity for nuclear scattering at these Bragg spots is $O(10^4)$ the predicted magnetic intensity. Muon spin-resonance (μ-SR) would be a possible probe, but the magnetic field from the current pattern in Fig. 2 is zero at most symmetry points and along the principal symmetry lines, where muons are known to sit preferentially. Perhaps, an

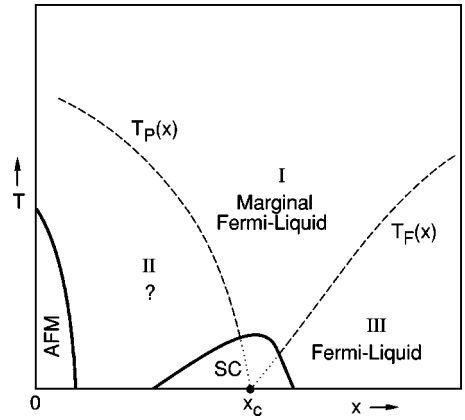

FIG. 1. Generic phase diagram of the cuprates for hole doping.

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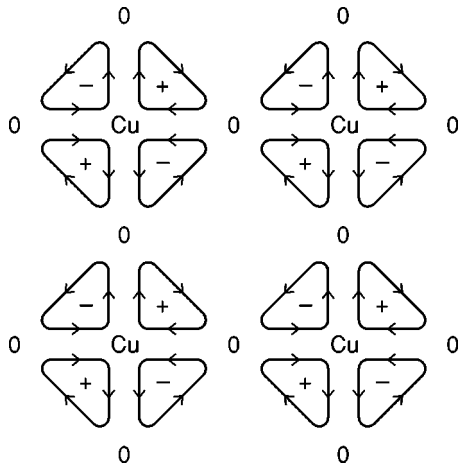

FIG. 2. Current pattern predicted in phase II of Fig. 1 in Refs. 5 and 6.

additional perturbation such as an external magnetic field can be used to lower the symmetry at the sites preferred by muons. In that case μ-*SR* could be used to search for the predicted phase.

I propose here an experiment which is a microscopic analog of circular dichorism. The idea is that angle-resolved photoemission spectroscopy (ARPES) at a specific point near the Fermi surface should have an electron yield which is different for right circularly polarized and left circularly polarized photons if the ground state has *T* breaking of the form shown in Fig. 2. Furthermore, the relative intensity should change in a systematic fashion with the momentum around the Fermi surface.

I present below the results of the calculation based on this idea and then discuss the feasibility of the experiment. The idea itself is more general than the specific application to copper oxides. Any time-reversal breaking phase will in general yield a different current density for right and left circularly polarized photons. (But the characteristic signature of the state, revealed by the momentum dependence of the asymmetry in the current for the left and right circular polarizations, must be calculated anew for each possibility.) The experiment may for example be tried to see if the superconducting state of the compound Sr_2RuO_4 (Ref. 7) breaks time-reversal symmetry.

II. ARPES WITH POLARIZED PHOTONS

My object is to deduce the polarization and symmetry dependence of ARPES current and a rough estimate of its magnitude. For this purpose, a simple calculation using tight-binding wave functions in the solid is sufficient.

Assume that a photon of energy ω shone on the crystal produces a free electron with momentum $\mathbf{p}$ and energy $E_{\mathbf{p}}$ at the detector due to absorption of the photon by an electronic state $|\mathbf{k}\rangle$ inside the crystal of the crystal momentum $\mathbf{k}$. The momentum of the photon is assumed very small compared to $\mathbf{k}$ and $\mathbf{p}$. The current $J_{\mathbf{p},\mathbf{k}}$ collected at the detector for uniform illumination over a given area is[8]

$$J_{\mathbf{p},\mathbf{k}}=2\pi e f(\epsilon_{\mathbf{k}})|\langle \mathbf{p}|H'|\mathbf{k}\rangle|^2\delta(E_{\mathbf{p}}-\epsilon_{\mathbf{k}}+\hbar\omega), \qquad (1)$$

where $f(\epsilon_{\mathbf{k}})$ is the Fermi function.

The primary contribution of the current is from the matrix element

$$\langle \mathbf{p}|H'|\mathbf{k}\rangle=\frac{-ie}{2mc}\int d\mathbf{r}\, e^{i\mathbf{p}\cdot\mathbf{r}}\mathbf{A}\cdot\nabla\Psi_{\mathbf{k}}(r), \qquad (2)$$

where $\mathbf{A}$ is the vector potential of the incident photons and $\Psi_{\mathbf{k}}(\mathbf{r})$ is the wave function of the state $|\mathbf{k}\rangle$. There is a smaller contribution due to the gradient of the potential at the surface which is briefly discussed at the end.

A. Wave functions

The creation operator for the tight-binding wave functions for the conduction band of Cu-O metals (assumed to be a two-dimensional metal) for the case that the difference in energy of the Cu-$d_{x^2-y^2}$ level ϵ_d and the O-$p_{x,y}$ levels $\epsilon_{\mathbf{p}}$ is much less than their hybridization energy, and when the direct oxygen-oxygen hopping parameter t_{pp} is set to zero are

$$|\mathbf{k}_o\rangle=\frac{d_k^+}{\sqrt{2}}+i\left(\frac{s_x p_{kx}^+ + s_y p_{ky}^+}{\sqrt{2}s_{xy}}\right), \qquad (3)$$

where $s_{x,y}=\sin k_{x,y}a/2$ and $s_{xy}^2=\sin^2 k_x a/2+\sin^2 k_y a/2$. Spin labels have been suppressed.

d_k^+, $p_{kx,y}^+$ are, respectively, the creation operators for the basis wave functions

$$\phi_d(\mathbf{k})=\frac{1}{\sqrt{N}}\sum_i e^{-i\mathbf{k}\cdot\mathbf{R}_i}\phi_d(\mathbf{r}-\mathbf{R}_i),$$

$$\phi_{p_{x,y}}(\mathbf{k})=\frac{1}{2\sqrt{N}}\sum_{i,a_m} e^{-i\mathbf{k}\cdot\mathbf{R}_i}e^{-i\mathbf{k}\cdot\mathbf{a}_m/2}\phi_{p_{x,y}}\left(\mathbf{r}-\mathbf{R}_i-\frac{a_m}{2}\right), \qquad (4)$$

where $\phi_d(\mathbf{r}-\mathbf{R}_i)$ is the $d_{x^2-y^2}$ atomic orbital at the Cu site R_i and $\phi_{p_x}(\mathbf{r}-\mathbf{R}_i-a_x/2)$ is the p_x wave function at the oxygen site at $\mathbf{R}_i+a_x/2$, etc, $a_m=\pm a_x,\ \pm a_y$.

In the circulating current phase, the conduction band wave function is modified to[9]

$$|\mathbf{k}\rangle=(|\mathbf{k}_0\rangle+\theta_0|\mathbf{k}_1\rangle)/\sqrt{1+\theta_o^2 s_x^2 s_y^2}, \qquad (5)$$

where

$$|\mathbf{k}_1\rangle\simeq s_x s_y(s_y p_{kx}^+ - s_x p_{ky}^+)/s_{xy}\,. \qquad (6)$$

In Eq. (5) θ_0 characterizes the strength of the symmetry breaking.

B. Matrix elements and current

In order to evaluate the matrix element, Eq. (2), I write

$$\phi_d(\mathbf{r})=\psi_d(|\mathbf{r}|)\frac{(x^2-y^2)}{r^2}, \qquad (7)$$

$$\phi_{p_\mu}(\mathbf{r})=\psi_p(|\mathbf{r}|)\frac{\mu}{|r|};\quad \mu=x,y.$$

$\psi_d(|\mathbf{r}|)$ and $\psi_{p_\mu}(|r|)$ are characterized by a falloff distance a of the order of the atomic size. Then

$$\nabla \phi_{p_\mu}(\mathbf{r}) \approx \left[\frac{1}{a}\left(\frac{\hat{x}x+\hat{y}y+\hat{z}z}{|\mathbf{r}|}\right)\frac{\mu}{|r|}+\frac{\hat{\mu}}{|r|}\right]\psi_p(|r|), \tag{8}$$

$$\nabla \phi_d(\mathbf{r}) \approx \left[\frac{1}{a}\left(\frac{\hat{x}x+\hat{y}y+\hat{z}z}{|\mathbf{r}|}\right)\frac{(x^2-y^2)}{r^2}+\frac{2(x\hat{x}-y\hat{y})}{r^2}\right]\psi_d(|r|).$$

We need the (two-dimensional) momentum distribution of the wave functions in Eq. (2). For this purpose define

$$\int d\mathbf{r}\, e^{i(p_x x+p_y y)}\nabla_\nu \phi_d(\mathbf{r}) \equiv i f_d^\nu(p_x,p_y); \quad \nu = x,y. \tag{9}$$

Note that $f_d^x(p_x,p_y)$ can be written as the product of an odd function of p_x and an even function of p_y, etc. Similarly,

$$\int d\mathbf{r}\, e^{i(p_x x+p_y y)}\nabla_\nu \phi_{p_\mu}(r) \equiv f_{p_\mu}^\nu(p_x,p_y). \tag{10}$$

$f_{p_\mu}^\mu(p_x,p_y)$ is the product of an even function of p_x and an even function of p_y, whereas $f_{p_\mu}^\nu(p_x,p_y)$ is the product of an odd function of p_x and an odd function of p_y. The definitions in Eqs. (9) and (10) ensure that all the f's are real. The $f(p)$'s falloff for p is of the order of the inverse atomic size. Therefore for p's in the first or second Brillouin zone they are approximately constant.

In terms of these quantities, the matrix element in Eq. (2) is calculated. Consider the case of left and right circularly polarized photons with vector potentials $\mathbf{A}_l$ and $\mathbf{A}_r$, respectively

$$\mathbf{A}_{l,r} = A(\hat{x} \pm i\,\hat{y}). \tag{11}$$

Then a straightforward calculation leads, to leading order in θ_0, to

$$\langle \mathbf{p}|H'|\mathbf{k}\rangle_{l,r} = \left(\frac{e}{2\sqrt{2}mc}\right) A \sum_{G_x,G_y} \delta(\mathbf{p}-\mathbf{k}-\mathbf{G}) \times \{(\mathbf{R}_0(\mathbf{G},\mathbf{p},\mathbf{k}) \pm \mathbf{i}\mathbf{I}_0(\mathbf{G},\mathbf{p},\mathbf{k})) \tag{12}$$

$$+ \theta_0(\pm R_1(\mathbf{G},\mathbf{p},\mathbf{k}) + i\, I_1(\mathbf{G},\mathbf{p},\mathbf{k}))\}, \tag{13}$$

where $\mathbf{G}=(G_x,G_y)$ are the reciprocal vectors, and

$$R_0(\mathbf{G},\mathbf{p},\mathbf{k}) = f_d^x(\mathbf{p}) + [g(G_y,k_x,k_y)f_{px}^y(\mathbf{p}) + g(G_y,k_x,k_y)f_{py}^y(\mathbf{p})] \tag{14}$$

$$I_0(\mathbf{G},\mathbf{p},\mathbf{k}) = f_d^y(\mathbf{p}) - [g(G_x,k_x,k_y)f_{p_x}^x(\mathbf{p}) + g(G_y,k_x,k_y)f_{p_y}^x(\mathbf{p})] \tag{15}$$

$$R_1(\mathbf{G},\mathbf{p},\mathbf{k}) = \left[\sin^2\left(\frac{k_y a}{2}\right) g(G_x,k_x,k_y) f_{p_x}^x(\mathbf{p}) - \sin^2\left(\frac{k_x a}{2}\right) g(G_y,k_x,k_y) f_{p_y}^x(\mathbf{p})\right] \tag{16}$$

$$I_1(\mathbf{G},\mathbf{p},\mathbf{k},\mathbf{k}) = \left[\sin^2\left(\frac{k_y a}{2}\right) g(G_x,k_x,k_y) f_{p_x}^y(\mathbf{p}) - \sin^2\left(\frac{k_x a}{2}\right) g(G_y,k_x,k_y) f_{p_y}^y\right]. \tag{17}$$

In the above

$$g(r,k_x,k_y) = \frac{\cos(ra/2)}{\sqrt{\sin^2(k_x a/2)+\sin^2(k_y a/2)}}. \tag{18}$$

In the usual experimental geometry, the contribution from each $\mathbf{G}$ is selected separately. For a particular $\mathbf{G}$, the current with polarization l or r to first order in θ is

$$J_{l,r}(\mathbf{G},\mathbf{p}) = \frac{e^2}{8m^2c^2}[(R_0^2+I_0^2) \pm 2\theta(R_0 R_1 + I_0 I_1)] \tag{19}$$

so the relative asymmetry of the current,

$$\Xi(\mathbf{G},\mathbf{p}) \equiv (J_l - J_r)/\tfrac{1}{2}(J_l+J_r), \approx 8\theta_0(R_0R_1+I_0I_1)/(R_0^2+I_0^2). \tag{20}$$

III. DISCUSSION OF ARPES—ASYMMETRY

Equations (19) and (20) are the principal result of the calculation. It is worthwhile noting that for $G_x=G_y$, the asymmetry vanishes along the zone diagonal $k_x=k_y$ and is maximum for the zone boundaries $(k_x a=\pi, k_y a=0)$; $(k_x a=0, k_y a=\pi)$ with a smooth variation in between. Asymmetry patterns for other G's may be obtained from Eqs. (14)–(17).

In Ref. 6, θ_0 is estimated to be $O(10^{-1})(x_c-x)^{1/2}$ for $x \leqslant x_c$. So at $x_c - x \approx 5\times 10^{-2}$, the asymmetry is predicted to be $O(10^{-1})$, at $T \approx T_p(x)$.

The proposed experiment is to measure the ARPES current in underdoped samples for a fixed relative geometry of the incident photon beam, crystalline surface, and the detector to select a $\mathbf{p}$ and $\mathbf{G}$ and then simply switch the polarization of the incident photons, and measure the current again. The experiment should then be repeated for different $\mathbf{p}$ and $\mathbf{G}$. The effect should set in for temperatures $T \lesssim T_p(x)$ and have a momentum dependence predicted by Eq. (20) and Eqs. (14)–(17).

The principal difficulty of the experiment is the possible presence of domains of the CC phase. The domains consist of regions in which (θ_0) in the wave function (5) is replaced by $(-\theta_0)$. This leads to a mutual switching of the pattern of the current within the unit cells (and a current flow along the domain boundary). The effect calculated in Eq. (20) to linear order in θ_0 then averages to zero for equal number of the two kinds of domains in the surface area S from which the current is collected. If the characteristic domain size is D, an effect proportional to $(D/S)^{1/2}\ \theta_0$ is still to be expected.

Also, Eq. (1) yields asymmetry terms proportional to θ_0^2, which are not affected by the domains. However, these may be too small to be observable.

In the above, circularly polarized photons, with the plane of polarization along the surface of the crystal, have been considered. There is also an effect linear in θ_0 for photons linearly polarized along the normal to the surface due to the potential gradient at the surface $(\nabla V)_s$. This effect, proportional to $(\nabla V)_s^2$ changes sign for a given $G_x = G_y$, as p_x and p_y are interchanged in a *d*-wave-like fashion. Observation of this effect requires rotating the sample. It is also affected by domains.

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[9] I have picked a different gauge for the wave functions compared to Ref. 6, but one which is the most convenient for the calculation reported here. Also, compared to Ref. 6, a coefficient has been absorbed on the definition of θ_0. In Ref. 6 additional modifications to the wave functions are discussed, which are not considered here. They produce further corrections.