Detection and Implications of a Time-reversal Breaking State in Underdoped Cuprates

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Abstract

We present general symmetry considerations on how a Time-reversal breaking state may be detected by angle-resolved photoemission using circularly polarized photons as has been proposed earlier. Results of recent experiments utilizing the proposal in underdoped cuprates are analysed and found to be consistent in their symmetry and magnitude with a theory of the Copper-Oxides. These together with evidence for a quantum critical point and marginal Fermi-liquid properties near optimum doping suggest that the essentials of a valid microscopic theory of the phenomena in the cuprates may have been found.

A major problem in condensed matter physics in the past decade and a half has been the search for a microscopic theory of high temperature superconductivity and associated normal state anomalies [1]. The normal state properties which presage superconductivity imply the inapplicability of the quasiparticle concept and are well-described by the marginal Fermi-liquid phenomenology [2]. This prescribes scale-invariant fluctuations governed by a quantum-critical point (QCP). A change of symmetry in the normal state with doping is then expected. The crucial question is whether a state with broken symmetry indeed exists and what is its nature? A microscopic theory based on a general model of the Cuprate compounds predicts an elusive phase which breaks time-reversal symmetry, without changing the translational symmetry of the lattice [3]. It is characterised by an ordered pattern of currents spontaneously circulating in prescribed patterns in each unit cell in the underdoped cuprates. Angle-resolved photoemission experiments using
polarised light were suggested to detect such a phase \[4\]. Here the general symmetry considerations necessary for the experiment and its analysis are derived. These results have been used in recent experimental work \[3\] to detect a time-reversal breaking phase in underdoped cuprates. The experiments are analysed here to show that the symmetry of the effect is characteristic of the class of the predicted phases and to rule out some other possibilities.

The current patterns in the ground state predicted by the microscopic theory \[3\] are illustrated in Fig. (1). Both arise from the same microscopic Hamiltonian; one or the other has lower energy depending on the detailed parameters of the model. It was suggested that the so called ”pseudogap phase” observed in these compounds is such a T-breaking phase.

The lack of a new translational symmetry in this phase makes it very hard to detect. Such a phase can be detected by measuring the difference in intensity of angle-resolved photoemission spectra (ARPES) for right and left circular polarized photons \[4\] in a mono-domain sample and analyzing the symmetry of the difference. The difference should set in below the characteristic pseudogap temperature.

For molecules absorbed on surfaces, a geometric effect has been derived, \[6\] which even without T-breaking yields intensity which depends on the circular polarization in ARPES experiments. Here we first generalise the geometric effect for the symmetries of a crystal and then derive the conditions necessary to distinguish the geometric effect from the effect due to T-breaking. Moreover, the symmetry of the experimental results as the direction of momentum of the outgoing electrons varies is shown to distinguish between different possible T-breaking phases.

**General Results**: Suppose a beam of photons of energy \(\omega\) shone on a crystal in the direction \(\hat{n}\) produces free-electrons with momentum \(\mathbf{p}\) and energy \(E_p\) at the detector. Let \(|\mathbf{k}\rangle\) denote the states of the crystal. Here \(\mathbf{k}\) is the wave-vector in the first Brillouin zone. Assuming the momentum of the photons is small compared to \(\mathbf{k}, \mathbf{p}\), the current \(J_p\) is given by

\[
J_p = 2\pi e \sum_{\mathbf{k}} f(\epsilon_k) |\langle \mathbf{p}|\mathbf{M}|\mathbf{k}\rangle|^2 \delta(E_p - \epsilon_k + \omega).
\]

(1)

where the matrix element is given by
\[ \langle p|M|k \rangle = \frac{-ie}{2mc} \int dr \Phi_p(r) A \cdot \nabla \psi_k(r). \quad (2) \]

and the summation is restricted by momentum conservation between \( p, k \) modulo the reciprocal vectors. Also \( A \) is the vector potential and \( \Phi_p(r) \) is the wave-function of the outgoing photoelectron of momentum \( p \). We may distinguish the two circular polarizations by \( A_{\ell,r} \)

\[ A_{\ell,r} = A_0(-\hat{x}' \pm i\hat{y}'), \quad (3) \]

where \( \hat{x}' \) and \( \hat{y}' \) are perpendicular to \( \hat{n} \), and the two matrix elements by \( M_{\ell,r}(k, p) \). We will assume that the crystal being studied is two-dimensional so that \( k \) refers to the momentum in the x-y plane. Note that, since the momentum of the photon is assumed negligible, \( p = (k + G) \). So, when \( p \) is in the mirror plane of the crystal, so is \( k \). (The converse is not true.) Let \( \hat{m} \) be the set of mirror planes of the crystal normal to the surface of the crystal. For reasons that will be clear shortly, we will consider only the situation in which \( \hat{n} \) lies in one of the \( \hat{m} \)-planes.

We may write in general that

\[ |p\rangle = \alpha_m|p, e\rangle + \beta_m|p, o\rangle \quad (4) \]

where under reflection \( R \) about a given \( m \)-plane,

\[ R_m|p\rangle = \alpha_m|p, e\rangle - \beta_m|p, o\rangle. \quad (5) \]

In Eq. (4), the eigenstates are divided into two parts, one of which has a real space representation even in reflection and the other has a real space representation odd in reflection about the given \( m \)-plane. Also because \( \hat{n} \) is contained in the mirror plane \( \hat{m} \)

\[ R_m^{-1}(A_{\ell}.\nabla)R_m = (A_{\ell}.\nabla) \quad (6) \]

Consider the group of the crystal wavefunctions \( |k\rangle \). In general, we may write,

\[ |k\rangle = \mu_m|k, e\rangle + \nu_m|k, o\rangle \quad (7) \]

so that in reflection about the \( m \)-plane,
\[ \mathcal{R}_m|k\rangle = \mu_m|k, e\rangle - \nu_m|k, o\rangle. \]  

(8)

In Eq. (7), the division of the wavefunctions follows the same convention as in Eq. (4). When T-symmetry is preserved, \( \nu_m = 0 \) if \( k \) lies in the mirror plane.

We are now finally ready to relate the matrix element for left circular polarization (lcp) with that for right circular polarization (rcp). Using Eq. (6), we can write,

\[ M_\ell = \langle p|\mathcal{R}^{-1}(A_r, \nabla)\mathcal{R}|k\rangle. \]  

(9)

Then using Eqs. (5) and (8),

\[ D_m \equiv |M_\ell|^2 - |M_r|^2 = 4\mathcal{R} \left( \alpha_m^* \beta_m |\mu_m|^2 \langle p, e|M_r^*|k, e\rangle \langle k, e|M_r|p, o\rangle \right. \]
\[ \left. + \beta_m^* \alpha_m |\nu_m|^2 \langle p, o|M_r^*|k, o\rangle \langle k, o|M_r|p, e\rangle \right. \]
\[ \left. + \mu_m \nu_m^* |\alpha_m|^2 \langle p, e|M_r^*|k, e\rangle \langle k, e|M_r|p, o\rangle \right. \]
\[ \left. + \nu_m \mu_m^* |\beta_m|^2 \langle p, o|M_r^*|k, o\rangle \langle k, o|M_r|p, e\rangle \right). \]  

(10)

The difference in \( J_p \) due to rcp and lcp follows through Eq. (1). In Eq. (10), \( \mathcal{R} \) picks up only the real part of its argument. We now separately consider the cases, T-symmetry preserved and T-symmetry broken.

**T-Symmetry preserved** As mentioned above a finite \( D \) exists even in this case due to the geometry of the experiment. For a T-preserving hamiltonian in a crystal with center of inversion \( \alpha, \beta, \mu, \nu \) may be taken real. For the geometric effect to be non-zero, it is necessary that either the state \( |p\rangle \) or the state \( |k\rangle \), does not have definite parity under the indicated reflection; this requires that three of the four quantities \( \alpha, \beta, \mu, \nu \) are non-zero. Since if \( p \) lies in a mirror-plane, so does \( k \), the former ensures the latter. Thus the geometric effect is zero if \( p \) lies in the plane \( \hat{m} \). Note that we assumed above that \( \hat{n} \) lies in the mirror plane \( \hat{m} \). If the experimental geometry is such that \( \hat{n} \) does not lie in the plane \( \hat{m} \), it is possible to show that the geometric effect is present in general even if \( p \) lies in the plane \( \hat{m} \). (The geometric effect is also zero, even if the above condition is satisfied, if the matrix elements in the product (10) are zero due to some other symmetries.)

The induced geometric effect must be distinguished in experiments from the proposed effect due to T-breaking. Towards this end, an important result following from Eq.(10) is that the geometric effect is odd with respect to reflection of \( p \) about the m-plane. Thus
if the outgoing plane wave with momentum \( p \) has a component \( \delta p_{\text{perp}} \) normal to this plane, (i.e. when \( \beta \neq 0 \), the difference of the intensity for \( \text{rcp-ARPES} \) and \( \text{lcp-ARPES} \) changes sign for \( \delta p_{\text{perp}} \to -\delta p_{\text{perp}} \).]

**T-symmetry Broken:** A complex hamiltonian breaks T-symmetry if no unitary transformation can convert it to a real form. Correspondingly, its eigenstates cannot be transformed to a real form by any gauge transformation. In translationally invariant media, broken T-symmetry implies broken inversion symmetry (provided Charge conjugation symmetry exists). In crystalline solids this is not necessary. A broken T-symmetry may or may not imply a broken reflection symmetry about some crystalline mirror plane \( \overline{m} \). Since inversion is a product of reflections about three mutually orthogonal mirror planes, inversion symmetry may be preserved while T-symmetry is broken. The specific proposals for T-symmetry breaking in copper-oxide metals that have been considered all lead to a broken reflection symmetry about one or more of the crystalline mirror planes.

The broken reflection symmetry about a given mirror plane \( \overline{m} \) attending a broken T-symmetry must be distinguished from that due to a structural or electronic distortion. In the latter cases, diffraction experiments, sensitive to density variations, detect the effect. For broken T-symmetry alone, the charge density retains the reflection symmetry about \( \overline{m} \) while the wavefunctions may not. (Specific examples of this will be given below.) In that case we will continue to call \( \overline{m} \) a mirror plane. For example in the copper-oxide lattice the \( x = 0, y = 0 \) and \( x = \pm y \) will continue to be called mirror planes even though due to broken T-symmetry, the wavefunctions may not be eigenstates of \( \Re \) about one or more of these planes.

This has the following consequence in Eq. (10). Consider \( p \) in a mirror plane \( \overline{m} \), so that \( |p\rangle = |p_e\rangle \). Then although \( k = p \) lies in the plane \( \overline{m} \) the wave-function \( |k\rangle \) has besides the usual component \( |k, e\rangle \), a component \( \theta|k, o\rangle \). It then follows that the third term in Eq. (10) is not zero for \( p, k \) in the plane \( \overline{m} \). This is true only for the mirror planes \( \overline{m} \) about which reflection symmetry is broken due to T-breaking. It then also follows that \( D_{\overline{m}} \) has a part which is even about the mirror planes \( \overline{m} \), as may also be checked from Eq. (10).

The above is fairly general. There is no reason why for a specific experimental geom-
etry and $p$. Eq. (10) may not be zero. The effect also has additional symmetries under rotation of $p$. They depend on details of the current pattern in the proposed state and must be examined separately for each proposal.

**Polarized ARPES for the Proposed State:** We will now consider the special T-breaking states [3] predicted for under-doped cuprates. Such states have been derived for a general Hamiltonian in the space of three orbitals per unit cell for non-local interactions above a critical value depending on the deviation of electronic density $x$ away from half-filling. The phase diagram in the $T - x$ plane, for the proposed T-breaking phase is consistent with the observed ”pseudo-gap” phase in the cuprates [4].

For the case that the difference in energy of the Cu-$d_{x^2-y^2}$ level $\epsilon_d$ and the O-$p_{x,y}$ levels $\epsilon_p$ is much less than their hybridization energy $t_{pd}$ and for the direct Oxygen-Oxygen hopping parameter $t_{pp} \ll t_{pd}$, the conduction band wave function in a tight-binding representation, without T-breaking may be written in terms of the ”anti-bonding” orbitals and the ”non-bonding” orbitals as follows:

$$|k\rangle \simeq (N_k)^{-1}[a_k^+ + 4t_{pp} s_x s_y (s_x^2 - s_y^2)n_k^+]|0\rangle,$$

where the anti-bonding and the non-bonding orbitals are created respectively by

$$a_k^+ = \frac{d_k^+}{\sqrt{2}} + \left( \frac{s_x p_{kx}^+ + s_y p_{ky}^+}{\sqrt{2s_{xy}}} \right), \quad n_k^+ = \left( s_y p_{kx}^+ - s_x p_{ky}^+ \right) / s_{xy}. \quad (12)$$

where $s_{x,y} = \sin(k_{x,y}a/2)$, $c_{x,y} = \cos(k_{x,y}a/2)$ and $s_{xy}^2 = \sin^2 \frac{k_{x,a}}{2} + \sin^2 \frac{k_{y,a}}{2}$ and $\epsilon_k = 2t_{pd}s_{xy}$. Spin labels have been suppressed. $d_k^+$, $p_{kx,y}^+$ are respectively the creation operators in momentum space for the $d_{x^2-y^2}$ atomic orbital at the Cu-site $R_i$ and the $p_{x,y}$ orbitals at the oxygen site at $(R_i + \frac{a x,y}{2})$, in each cell $i$.

If one approximates $|p\rangle$ by a plane wave $\sim \exp(ip \cdot r)$, the difference of the current from rcp ARPES and lcp ARPES, Eq. (10) vanishes for all $p$. This was the result presented in Ref. ([4], [8]). But for better outgoing wavefunctions which include the lattice and surface potentials, Eq. (10) is in general finite, except when $p$ lies in the plane $\hat{m}$, as shown above. For $p$ about this condition, the difference is odd, again as shown above.

Two sets of wavefunction for a T-breaking phase preserving translational invariance and inversion can be derived in the mean-field approximation [3]. The ground state of $|\Theta_1\rangle$ is made up of products of $|k, \theta_1\rangle$:
\[ |k, \theta_i \rangle = (N_k)^{-1}[a_{k, \theta_i}^+ + 4 \frac{t_{pp}}{\epsilon_k} s_x s_y (s_x^2 - s_y^2) n_k^+] |0 \rangle \]

\[ \theta_i = \pm \sum_k [s_x \langle p_{xk}^+ d_k \rangle - s_y \langle p_{yk}^+ d_k \rangle]. \]

\[ a_{k, \theta_i}^+ = \frac{d_k^+}{\sqrt{2}} + \left( \frac{s_x (1 + i \theta_i) p_{xk}^+ + s_y (1 - i \theta_i) p_{yk}^+}{\sqrt{2} s_{xy}} \right), \]

The ground state of \(|\Theta_2\rangle\) is made up of products of

\[ |k, \theta_2 \rangle = (N_k)^{-1}[a_{k, \theta_2}^+ + 4 \frac{t_{pp}}{\epsilon_k} s_x s_y (s_x^2 - s_y^2) n_k^+] |0 \rangle \]

\[ \theta_2 = \pm \sum_k [c_x \langle p_{xk}^+ d_k \rangle \pm c_y \langle p_{yk}^+ d_k \rangle]. \]

\[ a_{k, \theta_2}^+ = \frac{d_k^+}{\sqrt{2}} + \left( \frac{(s_x + i \theta_2 c_x) p_{xk}^+ + (s_y \pm i \theta_2 c_y) p_{yk}^+}{\sqrt{2} s_{xy}} \right), \]

In (13,14), the expectation values are determined self-consistently and \((\theta_1, \theta_2) << 1\) are assumed. The derived additional terms, proportional to the \(\theta\)'s break T-invariance because the effective Hamiltonians, of which Eqs. (13,14) are eigenstates, cannot be made real by any unitary transformation. The ground state currents corresponding to |\(\Theta_1\rangle\) and |\(\Theta_2\rangle\) are shown in Figs. (1a) and (1b) respectively. |\(\Theta_1\rangle\) retains mirror symmetry about the \(x = 0, y = 0\) planes, but not about the mirror planes \((\bar{m}_1 : x = \pm y)\). On the other hand |\(\Theta_2\rangle\) does not retain mirror symmetry about the mirror planes \((\bar{m}_2 : x = 0 \text{ and } y = 0)\). Two of the 4 possible domains of \(|\Theta_2\rangle\) retain reflection symmetry about \(x = y\) but not about \(x = -y\) while the other two have the opposite behavior. However \(D\) can be shown to be zero in \(|\Theta_2\rangle\) for all of them at both \(k_x = \pm k_y\) due to the symmetry of the transfer integral among the two oxygen orbitals in each unit cell.

The symmetry of the states (13,14) has the following consequence for \(D\). The state |\(\Theta_1\rangle\) produces an effect in \(D\) of order \(\theta_1\) which is even about the \(x = \pm y\) mirror planes and zero effect at the \(x = 0, y = 0\) mirror planes. The state |\(\Theta_2\rangle\) produce an effect in \(D\) of order \(\theta_2\) which is even about the mirror planes \(x = 0\) and \(y = 0\). From Eq. (10) it follows that the effect changes sign at these two mirror planes (i.e. if it is positive at one, it is negative at the other) and have maximum absolute magnitude at \((k_x, k_y) = (\pm \pi, 0)\).

The effect is zero at the mirror planes \(x = \pm y\).

Together with the geometric effect, the effective mirror planes defined as the plane for \(D = 0\) therefore appear rotated compared to the crystalline mirror planes; The rotation
is in opposite directions for two mutually orthogonal crystalline mirror planes \( \hat{\mathbf{m}} \). Further \( \hat{\mathbf{m}} \) are the \( x = \pm y \) planes for the state \( |\Theta_I\rangle \) and the planes \( x = 0 \) and \( y = 0 \) for the state \( |\Theta_{II}\rangle \).

There are additional modifications of the wave-functions near the chemical potential in the CC phase derived in [3], which are not included above.

*Analysis of the Experiments:* Recent polarized ARPES experiments [5] to look for the predicted effect [4] give results which are consistent with T-breaking in the underdoped phase of the cuprates. In one set of experiments [5], the region of momentum at the edge of the first Brillouin zone near the point \( (\pi/a, 0) \) was investigated thoroughly with \( \hat{\mathbf{n}} \) normal to the Cu – O plane. In the absence of a pseudogap the difference in the current for rcp and lcp ARPES was found to be odd about this point in traveling along the edge of the zone. This serves as a check on the experimental set up. In underdoped samples with pseudogap, a difference, symmetric about this point was observed below the temperature of appearance of the pseudogap and none was seen above this temperatures. This result has been seen in several underdoped samples; overdoped samples do not show the effect.

Two other features of the results are especially noteworthy in relation to microscopic theory. An investigation of the Brillouin zone near \( (0, \pi/a) \) in a given crystal produced an effect in \( \mathcal{D} \) of opposite sign to that around \( (\pi/a, 0) \) [5]. In other words the mirror planes \( x = 0 \) and \( y = 0 \) are *effectively* rotated in opposite directions. Therefore according to the symmetry considerations above these experiments are consistent with the state \( |\Theta_{II}\rangle \). Secondly, the magnitude of the effect is independent of the energy in the range investigated, \( \sim 0.5 \text{eV} \). This is important because in the microscopic theory [3], T-breaking is produced in a three-band model, by admixing due to long-range interactions, the states of the conduction and valence bands of the non-interacting Hamiltonian. The magnitude of the effect is then essentially uniform over the entire conduction band. (This is not a Fermi-surface effect). Moreover the absolute magnitude of the effect, about 5%, is consistent with the expectations.

In another set of experiments [5], the condition that the \( \hat{\mathbf{n}} \) is in the mirror plane was followed only for investigation with outgoing electron momentum \( \mathbf{p} \) in the \( x = y \) mirror plane. Then, within the experimental errors no difference was observed between
rcp and lcp ARPES when the point at the Fermi-surface in the \((k_x = k_y)\) direction was investigated. This is also consistent with the proposed CC state \(|\Theta_{II}\rangle\).

We now inquire if any other symmetry breaking can produce the observed effects. It may be seen that lattice distortions of the tetragonal to orthorhombic type, while preserving translational symmetry, do not have the symmetry to produce the observed effect. It may in principle be produced by distortions of the basic two-dimensional unit cell to the shape of a parallelogram, so that the relevant mirror plane symmetries are lost. Careful investigation of lattice distortions as a function of temperature [5] have not revealed any nor have such distortions have been reported elsewhere.

The difference of ARPES intensity for rcp and lcp photons exists for any T-breaking phase, be it due to spin-order or orbital order. Antiferromagnetic order at \(Q = (\pi/a, \pi/a)\) would produce a phase breaking reflection symmetry about the \(x = \pm y\) planes and is therefore incompatible with the observations. It would produce zero-effect for outgoing momenta along the \(\bar{m}\) planes \(x = 0, y = 0\) and maximum at the planes \(x = \pm y\). Further the effect would be zero at the zone-edge in these directions. A proposal combining the staggered flux phase [10], with the idea of a Quantum critical point near optimum doping has recently been advanced as a possibility for the underdoped compounds with the name, ”D-density wave” [11]. This phase has the same symmetry as the above antiferromagnetic phase with regard to the ARPES experiment. Moreover both these phases break translational symmetry so that the Fermi-surface consists of four pockets in the \(\pi - \pi\) directions. This is contrary to the observations by ordinary (linearly polarized ) ARPES in the cuprates. More complicated magnetic or structural symmetry-breakings can be envisaged producing the observed effect. But they would have to have escaped notice in direct diffraction experiments.

We also note that an Anyon state [12] can also be detected by ARPES experiments. For such a state the effective rotation of all the crystalline planes would be in the same direction.

**Conclusions:** The existence of a quantum critical point in the phase diagram [13] of Copper-oxides near optimum doping suggested that the central feature to be understood in the cuprates is the symmetry of the underdoped state. In the ARPES experiments, an
effect has been discovered which purely on symmetry grounds (as well as on grounds of its energy independence) has been identified here to be consistent with a proposed state \( \text{\textsuperscript{8}} \). Such a state breaking Time-reversal invariance but not translational invariance is the property of a general copper-oxide model with long range interactions. It is of-course obvious that a state which does not break translational invariance but has ground state currents can only be the property of a model with atleast three orbitals per unit cell. Approximate solutions of the same model lead to essentially all \( \text{\textsuperscript{4}} \) the universal features of the phase diagram of the cuprates, including the marginal Fermi-liquid fluctuations in the normal state near optimum doping, the pseudogap phenomena at underdoping, the crossover to a Fermi-liquid at overdoping, the vertex for ”d-wave” pairing as well as the right energy scale and coupling constant for the high \( T_c \) of the cuprates.

**Acknowledgements:** We have benefitted enormously from discussions with A.Kaminsky and J.C.Campuzano on the experimental results at various stages of their experiment. and from discussions with Elihu Abrahams, Eugene Blount and Ashvin Vishwanath about the theoretical issues.
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unaccompanied by thermodynamic signatures of a phase transition.
FIG. 1. Current patterns for the predicted T-breaking states that preserves translational invariance.