Some Consequences of $\mathcal{PT}$ Symmetry for Optical Rotation Experiments

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We perform a general symmetry analysis of optical experiments on samples in the "$\mathcal{PT}$ state," that is, samples for which 3D inversion symmetry $\mathcal{P}$ and time inversion symmetry $\mathcal{T}$ are each broken, but which are invariant under the product $\mathcal{PT}$. We show that $\mathcal{PT}$ symmetry is compatible with all known results on optical rotation in the high-temperature superconductors. We also find a *unique* and accessible experimental signature for the $\mathcal{PT}$ state.

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A number of optical experiments [1-4] have been carried out on various samples of high-temperature superconductors (HTSC), in search of signs of spontaneously broken time-reversal ($\mathcal{T}$) symmetry. The results of these experiments appear to support both positions on this question—that there is, or is not, such spontaneous symmetry breaking in the HTSC—since, of two sets of experiments specifically constructed to select only nonreciprocal effects, one [2] gave a null result, while another set [1,4] gave unambiguous positive results. A third set of experiments [3] strongly suggested broken $\mathcal{T}$; these experiments were not, however, constructed in such a way as to rule out reciprocal effects.

A recent paper by Dzyaloshinskii [5] has offered the possibility of resolving nearly all the known experimental results. Dzyaloshinskii pointed out the consequences of a hypothesized "antiferromagnetic" configuration of the HTSC, in which $\mathcal{T}$ is broken in each of the metallic CuO planes, but the *sign* of the order parameter strictly alternates from one plane to the next. If there is only one such plane per chemical unit cell, the resulting material is both $\mathcal{T}$ and $\mathcal{P}$ invariant (where $\mathcal{P}$ is 3D inversion), and no bulk effects of the broken symmetry can be expected. If instead there are two planes per unit cell, then the material breaks *both* $\mathcal{T}$ and $\mathcal{P}$, while the combined operation $\mathcal{PT}$ remains a good symmetry. We call this state the "$\mathcal{PT}$ state" [6]. We note that there are good reasons to believe that, if $\mathcal{T}$ is spontaneously broken in each plane, the planes will order with alternating sign, i.e., antiferromagnetically [7].

Dzyaloshinskii showed that a material in the $\mathcal{PT}$ state has in general nonzero magnetoelectric coefficients of linear response, and that such coefficients give rise to a (complex) rotation of the polarization of light upon reflection. He also stated that $\mathcal{PT}$ invariance *forbids* optical rotation in transmission.

It follows that an assumption of the $\mathcal{PT}$ state for the HTSC is consistent with many of the experimental results: the null result of Spielman *et al.* [2] seeking optical rotation in transmission, the positive results of Lyons *et al.* [1] for reflection, and the positive results of Weber *et al.* [3] in reflection. The idea gained further credit when another prediction of the $\mathcal{PT}$ model was tested. Lyons [4] looked for rotation in reflection from samples of La_2CuO_4, which, unlike the other HTSC materials tested, has a single conducting plane per unit cell. In accord with the prediction of the $\mathcal{PT}$ model, Lyons found a null result for the 2:1:4 material.

There remains one outstanding result which appears to be inconsistent with the $\mathcal{PT}$ state for the HTSC, namely, the optical rotation in transmission found for $Bi_2Sr_2CaCu_2O_8$ (2:2:1:2) by Weber *et al.* [3]. Because the $\mathcal{PT}$ model reconciles all the other results (including one which was predicted by it), we feel it is worthwhile to pursue further the implications of the $\mathcal{PT}$ state. In the following analysis we show that the rotation in transmission seen by Weber *et al.* is consistent with the $\mathcal{PT}$ state, thus reconciling all known experimental results within the $\mathcal{PT}$ model. We also propose an experimental test which can uniquely distinguish the $\mathcal{PT}$ state from other broken-symmetry states.

Symmetry analysis.— We consider idealized experiments in which polarized light is incident on a sample from either the right (r) or left (l) side, and the resulting reflected and transmitted light is detected. We take all outgoing (**o**) waves to be linear functions of the incoming (**i**) waves:

$$\mathbf{o}=\mathbf{S}\cdot\mathbf{i}\,. \tag{1}$$

We choose as a basis the electric fields of circularly polarized (cp) light: l^o_+ is the outgoing wave on the left side of (+) cp, etc. Thus

$$\mathbf{o}=\begin{pmatrix} l_+ \\ r_+ \\ l_- \\ r_- \end{pmatrix}^o , \tag{2}$$

and similarly for **i**. The scattering matrix **S** is then

$$\mathbf{S}=\begin{pmatrix} \begin{pmatrix} R^l_{++} & T^r_{++} \\ T^l_{++} & R^r_{++} \end{pmatrix} & \begin{pmatrix} R^l_{+-} & T^r_{+-} \\ T^l_{+-} & R^r_{+-} \end{pmatrix} \\ \begin{pmatrix} R^l_{-+} & T^r_{-+} \\ T^l_{-+} & R^r_{-+} \end{pmatrix} & \begin{pmatrix} R^l_{--} & T^r_{--} \\ T^l_{--} & R^r_{--} \end{pmatrix} \end{pmatrix} . \tag{3}$$

We now ask how the matrix $\mathbf{S}$ transforms under $\mathcal{P}$ and $\mathcal{T}$. For $\mathcal{P}$ we reason as follows. Any experiment describable by Eq. (1) implies that an inverted experiment, in which inputs, outputs, and sample are subjected to $\mathcal{P}$, is also possible, since the laws of electromagnetism do not violate parity [8]. Thus we get $\hat{\mathcal{P}}\mathbf{o}=\hat{\mathcal{P}}\mathbf{S}\hat{\mathcal{P}}^{-1}\hat{\mathcal{P}}\mathbf{i}$, so that $\hat{\mathcal{P}}\mathbf{S}\hat{\mathcal{P}}^{-1}$ is the appropriate scattering matrix for the space-inverted sample.

$\mathcal{P}$ exchanges left and right sides, leaves the cp unaltered, and reverses the wave vector $\mathbf{k}$ of the light; thus

$$\hat{\mathcal{P}}\begin{pmatrix} l+ \\ r+ \\ l- \\ r- \end{pmatrix}^{i,o} = \begin{pmatrix} r+ \\ l+ \\ r- \\ l- \end{pmatrix}^{i,o} \tag{4}$$

so that

$$\hat{\mathcal{P}}=\begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}=\begin{pmatrix} \sigma_x & \mathbf{0} \\ \mathbf{0} & \sigma_x \end{pmatrix}=\hat{\mathcal{P}}^{-1}, \tag{5}$$

where σ_x is the 2×2 Pauli matrix. Thus we get

$$\hat{\mathcal{P}}\mathbf{S}\hat{\mathcal{P}}^{-1}=\begin{pmatrix} \begin{pmatrix} R^r_{++} & T^l_{++} \\ T^r_{++} & R^l_{++} \end{pmatrix} & \begin{pmatrix} R^r_{+-} & T^l_{+-} \\ T^r_{+-} & R^l_{+-} \end{pmatrix} \\ \begin{pmatrix} R^r_{-+} & T^l_{-+} \\ T^r_{-+} & R^l_{-+} \end{pmatrix} & \begin{pmatrix} R^r_{--} & T^l_{--} \\ T^r_{--} & R^l_{--} \end{pmatrix} \end{pmatrix}. \tag{6}$$

For time reversal $\mathcal{T}$ we cannot make a strictly analogous argument to that used for $\mathcal{P}$, the reason being the second law of thermodynamics. The strict time reverse of an experiment with dissipation is an experiment with "antidissipation," which is not realizable. In the past, the "principle of reciprocity" (POR) has been invoked to analyze time-reversed light-scattering experiments [9]. The POR relates *intensities* at polarizing analyzers in an experiment and its time-reversed version. Since we are interested in the phase as well as the magnitude of the elements of the scattering matrix $\mathbf{S}$, we will avoid invoking the POR.

Instead, we can generalize an argument due to Halperin [10]. Consider an emitter at z_1 emitting cp light, so that the emitted electric field is $\mathbf{E}(z_1)=(1/\sqrt{2})(\hat{\mathbf{x}}\pm i\hat{\mathbf{y}})\equiv\mathbf{E}_\pm(z_1)$ (time dependence $e^{-i\omega t}$ is implicit). In linear response the field at z_2 is

$$E_\alpha(z_2)=\sum_\beta \chi_{\alpha\beta}E_\beta(z_1), \tag{7}$$

where the functions $\chi_{\alpha\beta}=\chi_{\alpha\beta}(z_2,z_1,\omega,\Phi)$ are the linear response functions of the "system," i.e., the sample and the surrounding medium, and (α,β) are *Cartesian* coordinates x or y. Φ is an order parameter which is nonzero when the sample breaks $\mathcal{T}$. Finally, we define $E_{\mu\nu}(z_2,z_1,\omega,\Phi)$ ($\mu,\nu=+$ or $-$) to be the amplitude of μ-cp electric field at position z_2, given that a source is emitting ν-cp light at z_1 of unit amplitude. It is easily shown [10] that

$$E_{++}(z_2,z_1,\omega,\Phi)=\tfrac{1}{2}\chi_+(z_2,z_1,\omega,\Phi) \tag{8}$$

and similarly $E_{--}=\frac{1}{2}\chi_-$, where

$$\chi_\pm=\chi_{xx}+\chi_{yy}\pm i(\chi_{xy}-\chi_{yx}). \tag{9}$$

We now define

$$\bar{E}_{++(--)}(z_2,z_1,\omega)\equiv E_{++(--)}(z_2,z_1,\omega,-\Phi),$$

that is, $\bar{E}$ gives the response of the time-reversed sample; thus $\bar{E}_{\pm\pm}=\frac{1}{2}\overline{\chi_\pm}$. The $\chi_{\alpha\beta}$, being coefficients of linear response, obey Onsager relations [11] as follows:

$$\chi_{\alpha\beta}(z_2,z_1,\omega,-\Phi)=\chi_{\beta\alpha}(z_1,z_2,\omega,\Phi). \tag{10}$$

This gives $\overline{\chi_\pm}(z_2,z_1,\omega)=\chi_\mp(z_1,z_2,\omega)$, or

$$\bar{E}_{++(--)}(z_2,z_1,\omega)=E_{--(++)}(z_1,z_2,\omega). \tag{11}$$

A similar derivation gives $\bar{E}_{+-}(z_2,z_1)=E_{+-}(z_1,z_2)$ and $\bar{E}_{-+}(z_2,z_1)=E_{-+}(z_1,z_2)$.

We can make contact with our earlier notation by simply strategically locating z_1 and z_2 to give reflection or transmission from either side. The above results for the $\bar{E}_{\mu\nu}$ then give us the entries in the matrix $\bar{\mathbf{S}}\equiv\mathbf{S}(-\Phi)$ in terms of those $\mathbf{S}$. Thus we get

$$\bar{\mathbf{S}}=\mathbf{S}(-\Phi)$$
$$=\begin{pmatrix} \begin{pmatrix} R^l_{--} & T^l_{--} \\ T^r_{--} & R^r_{--} \end{pmatrix} & \begin{pmatrix} R^l_{+-} & T^l_{+-} \\ T^r_{+-} & R^r_{+-} \end{pmatrix} \\ \begin{pmatrix} R^l_{-+} & T^l_{-+} \\ T^r_{-+} & R^r_{-+} \end{pmatrix} & \begin{pmatrix} R^l_{++} & T^l_{++} \\ T^r_{++} & R^r_{++} \end{pmatrix} \end{pmatrix}. \tag{12}$$

We can rewrite the above in the simple form $\bar{\mathbf{S}}=\hat{\jmath}\mathbf{S}^T\hat{\jmath}^{-1}$, where $\mathbf{S}^T$ is the transpose and

$$\hat{\jmath}=\begin{pmatrix} \mathbf{0} & \mathbf{1} \\ \mathbf{1} & \mathbf{0} \end{pmatrix}=\hat{\jmath}^{-1}. \tag{13}$$

Finally, for the $\mathcal{PT}$ state, we have that $\hat{\mathcal{P}}\bar{\mathbf{S}}\hat{\mathcal{P}}^{-1}=\mathbf{S}$. This gives

$$\mathbf{S}=\hat{\mathcal{P}}\hat{\jmath}\mathbf{S}^T\hat{\jmath}\hat{\mathcal{P}}$$
$$=\begin{pmatrix} \begin{pmatrix} R^r_{--} & T^r_{--} \\ T^l_{--} & R^l_{--} \end{pmatrix} & \begin{pmatrix} R^r_{+-} & T^r_{+-} \\ T^l_{+-} & R^l_{+-} \end{pmatrix} \\ \begin{pmatrix} R^r_{-+} & T^r_{-+} \\ T^l_{-+} & R^l_{-+} \end{pmatrix} & \begin{pmatrix} R^r_{++} & T^r_{++} \\ T^l_{++} & R^l_{++} \end{pmatrix} \end{pmatrix}. \tag{14}$$

We are now prepared to investigate the constraints imposed by $\mathcal{PT}$ symmetry on the rotation of the polarization of light. Consider an input beam of x-polarized light on the left: $\mathbf{i}=(1/\sqrt{2})(1\ 0\ 1\ 0)^T$. The transmitted wave is in general elliptically polarized. Define

$$\Psi_T^l = \frac{r_+^o}{r_-^o} = \frac{T_{++}^l + T_{+-}^l}{T_{-+}^l + T_{--}^l}\,; \qquad (15)$$

then the azimuth θ and ellipticity ϵ of the transmitted wave are given by [12] $\theta = -\frac{1}{2}\arg(\Psi)$ and $\tan\epsilon = (|\Psi|-1)/(|\Psi|+1)$. From Eqs. (3) and (14) we see (as noted by Dzyaloshinskii [5]) that $\mathcal{PT}$ symmetry alone is sufficient to give the constraint $T_{++}^i = T_{--}^i$ for $i = r,l$. However, T_{+-}^i and T_{-+}^i are *unconstrained*, so that we find no general constraints on θ or ϵ. The same conclusion holds for the reflected wave.

Let us then increase the symmetry of scattering system (sample). Our postulated $\mathcal{PT}$ symmetry for HTSC can be augmented by orthorhombic or tetragonal symmetry, giving the magnetic point groups [5,13] $m'm'm'$ or $4/m'm'm'$, respectively. For the orthorhombic case, m' (equal to reflection times $\mathcal{T}$) symmetry in the a-b plane gives $S^T = S$. (Here and henceforth we assume propagation along the c axis.) Given these additional constraints, we find, for $\hat{\mathbf{x}}$-polarized input, $\Psi_T^l = (\Psi_T^r)^{-1} \neq 1$. Thus, in the orthorhombic case, an input $(\hat{\mathbf{x}})$ which propagates *unchanged* in the absence of $\mathcal{T}$ and $\mathcal{P}$ breaking is subject to a rotation in the $\mathcal{PT}$ state.

Tetragonal symmetry is equivalent to full rotational invariance along the axis of propagation (for c-directed propagation). Imposing this further constraint sets the off-diagonal blocks of $\mathbf{S}$ (and hence of $\bar{\mathbf{S}}$ and $\hat{\mathcal{P}}\bar{\mathbf{S}}\hat{\mathcal{P}}$) to $\mathbf{0}$. It is then easily shown that, given $\mathcal{PT}$ symmetry and an arbitrary linearly polarized input, $\Psi_T^i/\Psi_{\text{in}} = T_{++}^i/T_{--}^i = 1$. Thus, *with the qualification of rotational invariance* we find "no optical rotation in transmission" for the $\mathcal{PT}$ state.

For the case of reflection we still have that $R_{++}^i \neq R_{--}^i$ in general. Thus both rotation and ellipticity may result from reflection from a sample of arbitrary rotational symmetry in the $\mathcal{PT}$ state, as suggested by Dzyaloshinskii.

Anisotropic dissipation.—The fact that $R_{++}^i \neq R_{--}^i$ is a signature usually associated with broken $\mathcal{T}$ symmetry; as we have seen, it is also associated with broken $\mathcal{P}$ and $\mathcal{T}$, with $\mathcal{PT}$ unbroken. We find another signature of the $\mathcal{PT}$ state below, which is usually associated with broken $\mathcal{P}$. We will continue to work with the assumption of rotational symmetry; the consequences of dropping this assumption will be discussed subsequently.

Define $(\gamma_+^l)^2 = |R_{++}^l|^2 + |T_{++}^l|^2$ so that $1-(\gamma_+^l)^2$ gives the absorption of + cp light incident from the left, and similarly for the others. If $\mathcal{P}$ is broken, we get

$$(\gamma_+^r)^2 - (\gamma_-^r)^2 = (\gamma_-^l)^2 - (\gamma_+^l)^2, \qquad (16)$$

which says that preferential absorption is sensitive only to the handedness (right or left) of the light—which is what we expect for broken $\mathcal{P}$. For broken $\mathcal{T}$ we get

$$(\gamma_+^r)^2 = (\gamma_+^l)^2 = \gamma_+^2, \quad (\gamma_-^r)^2 = (\gamma_-^l)^2 = \gamma_-^2, \qquad (17)$$

i.e., the preferential absorption is sensitive only to the absolute ($\pm$) circular polarization.

For broken $\mathcal{T}$ and $\mathcal{P}$ but unbroken $\mathcal{PT}$, we get

$$(\gamma_+^l)^2 - (\gamma_-^l)^2 = |R_{++}^l|^2 - |R_{--}^l|^2,$$

$$(\gamma_+^r)^2 - (\gamma_-^r)^2 = |R_{++}^r|^2 - |R_{--}^r|^2 = |R_{--}^l|^2 - |R_{++}^l|^2,$$

so that, finally,

$$(\gamma_+^r)^2 - (\gamma_-^r)^2 = (\gamma_-^l)^2 - (\gamma_+^l)^2 \quad (\mathcal{PT}\text{ state}). \qquad (18)$$

We see that Eqs. (16) and (18) are identical: A $\mathcal{PT}$ material is like one with broken $\mathcal{P}$ alone in that its circular dichroism is sensitive only to the *handedness* of the incoming light.

Equation (18) can also be shown to hold for the orthorhombic case $(m'm'm')$. Furthermore, regardless of the degree of rotational symmetry, we know directly from Eq. (14) that

$$|R_{++}^r|^2 - |R_{--}^r|^2 = -(|R_{++}^l|^2 - |R_{--}^l|^2). \qquad (19)$$

Therefore, any experiment which can measure explicitly the right-hand side (RHS) and the left-hand side (LHS) on a single sample could be used to *unambiguously* signal the $\mathcal{PT}$ state, for which Eq. (19) is *uniquely* true: broken $\mathcal{T}$ alone gives

$$|R_{++}^r|^2 - |R_{--}^r|^2 = +(|R_{++}^l|^2 - |R_{--}^l|^2)$$

while broken $\mathcal{P}$ alone gives RHS=LHS=0.

To our knowledge at least one group [14] can now measure $|R_{++}^i|^2 - |R_{--}^i|^2$. A test of the $\mathcal{PT}$ state for the HTSC, using Eq. (19), would then require a nearly-defect-free sample, so that the antiferromagnetic ordering is maintained from one surface to the other. Ideally, the size of broken $\mathcal{T}$ *domains* within the planes should also be larger than the laser spot size.

We have defined the $\mathcal{PT}$ state, and examined its consequences for experiments on optical rotation. We find that optical rotation in transmission is forbidden in the $\mathcal{PT}$ state only if the additional requirement of rotational symmetry is imposed. The experiments of Weber *et al.* [3] on 2:2:1:2 show large optical rotation in transmission, which is not allowed if we assume the $\mathcal{PT}$ model plus tetragonal symmetry for the material. In fact, 2:2:1:2 is only pseudotetragonal [14], as there is usually an incommensurate modulation of the lattice along the a or b axis. The resulting symmetry is orthorhombic, which we have shown to give a rotation [15] in the $\mathcal{PT}$ state, even for input which is polarized along a principal symmetry axis of the material. The experiment of Spielman *et al.* [2] measures $\arg(T_{--}^l/T_{++}^r)$. Interestingly, this quantity may be nonzero for the general $\mathcal{PT}$ state, but is strictly zero for orthorhombic symmetry or higher—and hence for the HTSC in the $\mathcal{PT}$ state.

Our symmetry analysis has also led to a new relation [Eq. (19)] which uniquely distinguishes the $\mathcal{PT}$ state from other broken symmetries. This relation suggests a further test for the $\mathcal{PT}$ state which appears to be within the realm of experimental feasibility. Any material obeying Eq. (19) *must* be in the $\mathcal{PT}$ state, that is, it must break $\mathcal{P}$ and $\mathcal{T}$ while remaining invariant under $\mathcal{PT}$. Equation (19) holds without further assumptions—in particular, without assuming rotational invariance. It applies to a quantity which is currently experimentally accessible. We look forward to its future application as a test of the $\mathcal{PT}$ state for the HTSC.

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Note added.—Since this work was submitted, we have received a paper from Spielman *et al.* (to be published) reporting a null result in reflection for 1:2:3 and 2:2:1:2. This result disagrees with the $\mathcal{PT}$ model in the weak sense that a null result is found where a positive result is allowed. We also have received a theoretical paper by Shelankov and Pikus (to be published) which treats questions similar to those addressed in this paper. In particular, their analysis of the effect of time reversal on the scattering matrix agrees with ours.

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[15] Weber *et al.* (Ref. [3]) also observed that they could control the *sign* of the observed rotation by cooling their samples in a uniform magnetic field along the c axis. This implies that one type of antiferromagnetic order is favored over the other by a uniform field, which seems at least puzzling. While we do not have the solution to this puzzle, we note that many early experiments with antiferromagnetic, magnetoelectric insulators observed precisely the same phenomenon, i.e., biasing of *antiferromagnetic* domains by a uniform applied field. Apparently, the effect is still not understood. See, for example, G. T. Rado and V. J. Folen, Phys. Rev. Lett. **7**, 310 (1961); D. N. Astrov, Zh. Eksp. Teor. Fiz. **40**, 1035 (1961) [Sov. Phys. JETP **13**, 729 (1961)]; T. J. Martin and J. C. Anderson, Phys. Lett. **11**, 109 (1964).