Ellipsometry and broken time-reversal symmetry in the high-temperature superconductors

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We discuss ellipsometric experiments seeking evidence of broken time-reversal symmetry in the high-temperature copper oxide superconductors. We use both a generalized symmetry analysis, and a magnetoelectric model suggested by Dzyaloshinskii, to argue that the hypothesis that best fits the various experimental results is the $\mathcal{PT}$-invariance hypothesis—which assumes broken $\mathcal{T}$ symmetry in each plane, and antiferromagnetic (alternating) order of the broken symmetry in the c direction. We suggest two experimental tests of the $\mathcal{PT}$-invariant model; one of these is sufficient to rule out any other broken-symmetry state and so has the potential to be extremely useful.

I. INTRODUCTION

There exists a wide variety of theories of the normal state of the doped (metallic) phase of the high-temperature superconductors (HTSC). A subset of these theories[1] predicts that time-reversal ($\mathcal{T}$) symmetry is spontaneously broken in the HTSC. (This broken symmetry is distinct from that seen in the undoped phase, which has long-ranged antiferromagnetic order of the copper spins.) A common feature of these theories is the existence of spontaneous (charge or spin) currents in the planes, which circulate either right or left handed. Hence a simple, generic model for the broken $\mathcal{T}$ symmetry in the doped phase involves an order parameter for each plane that takes one of two discrete values (+ or −).

A number of experiments[2–8] have been performed in search of the putative broken $\mathcal{T}$ symmetry of the doped phase of the HTSC. In this paper we focus on those experiments seeking signs of broken $\mathcal{T}$ symmetry in the response of HTSC samples to polarized light.[9] An experiment with such aim, reported by Lyons *et al.*,[3] found circular dichroism (CD) in reflection from samples of $YBa_2Cu_3O_{7-x}$ (1:2:3) and $Bi_2Sr_2CaCu_2O_8$ (2:2:1:2). This experiment suggested broken $\mathcal{T}$ symmetry in these materials, but could not rule out CD arising from other effects such as lattice distortion. Subsequently, Spielman *et al.* conducted tests sensitive only to the nonreciprocal component of circular birefringence (CB) in transmission for 1:2:3 (Ref. 5) and for 2:2:1:2 (Ref. 6). A positive signal from such a test is conclusive evidence for broken $\mathcal{T}$ symmetry; the result was, however, null for this experiment. In contrast, Weber *et al.*[8] reported CD in reflection (and subsequently in transmission) from 1:2:3, and CB in transmission through 2:2:1:2. These experiments again strongly suggest broken $\mathcal{T}$ symmetry but are not conclusive. For instance, the transmission (CB) experiments involved measuring the rotation of light (initially polarized along a principal axis of the material to avoid effects from linear birefringence) after passing only one way through the sample, and thus could not distinguish reciprocal from nonreciprocal effects.

More recently, Lyons *et al.*[4] have reported a modification of their experiments that gives a signal sensitive only to $|R_{++}|^2-|R_{--}|^2$. [Here R_{++} is the amplitude for reflected +-circularly-polarized (CP) light, given that unit amplitude of +-CP light is incident; and similarly for R_{--}.] As shown by Halperin,[10] a nonzero value for this quantity is unambiguous evidence of broken $\mathcal{T}$ symmetry. Lyons *et al.* found a nonzero value for $|R_{++}|^2-|R_{--}|^2$ in many (but not all) samples of 1:2:3; they also report a null result for the same test on a single sample of $La_{2-x}Sr_xCuO_4$ (2:1:4). While the sample dependence of these results is puzzling, the positive results obtained indicate unambiguously that $\mathcal{T}$ symmetry is broken, both above and below T_c, at least in 1:2:3. This apparently conclusive result is however, challenged by recent results from reflection experiments by Spielman *et al.*[7] who found a null result for the quantity $\arg(R_{++}/R_{--})$ for both 1:2:3 and 2:2:1:2 samples.

We thus find that the question of broken $\mathcal{T}$ symmetry in the HTSC remains unresolved by the available experimental results. There are however some theoretical results that may help to resolve the question. Analyses of coupled planar systems (with $\mathcal{T}$ symmetry broken in each plane) by us[11] and by Rojo and Leggett[12] concluded that, under very general circumstances, antiferromagnetic order ($\cdots+-+-+\cdots$) of the symmetry breaking in the planes is preferred energetically. Simultaneous but independent work by Dzyaloshinskii[13] showed that antiferromagnetic (AFM) order of the planes, for 1:2:3 and for 2:2:1:2, would give $R_{++}\neq R_{--}$ but $T_{++}=T_{--}$, thus (apparently) allowing the null results of Spielman *et al.* in transmission to be reconciled with the positive results of Lyons *et al.* in reflection. Dzyaloshinskii also pointed out that 2:1:4 (with AFM order of the planes) falls in a different symmetry class, and should give a null result in reflection as well as transmission. This prediction was subsequently tested and confirmed by the Lyons experiment on 2:1:4 mentioned above.

Recently[14] we have performed a general symmetry analysis of transmission and reflection experiments for normal incidence. We find that this analysis allows reconciliation of all the above experimental results but one, by assuming broken $\mathcal{T}$ symmetry in each plane, with the planes ordered antiferromagnetically (Fig. 1). The outstanding result which is not predicted by this model is the null result of Spielman *et al.*[7] for $\arg(R_{++}/R_{--})$. The rotation in transmission seen by Weber *et al.*[8] was shown to be allowed by this model, even though the result of the Spielman *et al.*[5,6] transmission experiment is predicted to be null. Since, as pointed out by Dzyaloshinskii[13] (and as seen in Fig. 1), AFM order of the planes (for 1:2:3 and 2:2:1:2) breaks *three-dimensional* spatial inversion ($\mathcal{P}$) symmetry as well as time-reversal ($\mathcal{T}$) symmetry, but is invariant under the product $\mathcal{PT}$, we call this model the $\mathcal{PT}$-invariant model for the HTSC.

A symmetry analysis similar to ours has been performed by Shelankov and Pikus.[15] We find complete agreement where the two analyses overlap. The work of Shelankov and Pikus differs from Ref. 14 and the present work in that it considers the effects of time reversal on partially as well as fully-polarized light, but it does not consider the implications for experiment of the $\mathcal{PT}$-invariant state.

In this paper we offer a detailed discussion of the analysis of Ref. 14. In particular, we treat the question of time reversal as applied to light-scattering experiments in some detail, showing a connection between the "principle of reciprocity"[16,17] and the "Onsager" relations for linear-response coefficients.[18] We also present a concrete realization of the $\mathcal{PT}$-invariant model: We solve for the reflected and transmitted waves for light normally incident on a magnetoelectric slab or orthorhombic or tetragonal symmetry. Nonzero magnetoelectric coefficients[13,19] are a consequence of and a realization of $\mathcal{PT}$ symmetry, i.e., they are indicative of separately broken $\mathcal{P}$ and $\mathcal{T}$ symmetries but are under combined $\mathcal{PT}$ symmetry. Our solution for the magnetoelectric slab thus provides another route to the conclusions obtained from the pure symmetry analysis.

Finally, we discuss two experimental tests of $\mathcal{PT}$ symmetry that are suggested by our analysis. Each requires for its realization a sample for which a single domain is accessible from *both surfaces.* This is however the only obstacle to performing these tests; and, based on reported characteristics of single-crystal samples of $Bi_2Sr_2CaCu_2O_8$,[8] it seems that this obstacle is not insurmountable. Hence, we hope that this analysis might help in resolving the question of the existence and nature of broken time-reversal symmetry in the HTSC.

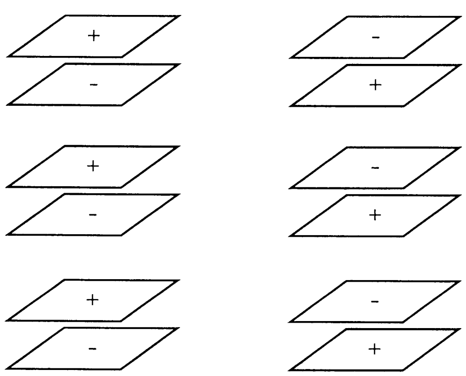

FIG. 1. The $\mathcal{PT}$-invariant model for the HTSC. We assume time-reversal ($\mathcal{T}$) symmetry is spontaneously broken in each metallic plane; the sign of the broken symmetry can be either + or −. The pictured configurations involve two planes per unit cell [appropriate for $YBa_2Cu_3O_{7-x}$ (1:2:3) and $Bi_2Sr_2CaCu_2O_8$ (2:2:1:2)], with strictly alternating signs of the broken $\mathcal{T}$ symmetry. The stack on the left is converted to the one on the right by *either* $\mathcal{T}$ *or* three-dimensional inversion $\mathcal{P}$. Since the two stacks are distinguishable, the model breaks $\mathcal{T}$ and $\mathcal{P}$ symmetries; but it is invariant under the combined $\mathcal{PT}$.

II. SYMMETRY ANALYSIS

We consider monochromatic, fully-polarized light at normal incidence on a perfect sample. A perhaps obvious but crucial ingredient of our analysis is a distinction between the two surfaces of the sample, which we label "left" (l) and "right" (r), respectively, (see Fig. 2). Hence our reflection and transmission coefficients take superscripts denoting the surface of incidence; for example, for unit amplitude +-CP light incident on the left, R^l_{++} and T^l_{++} give the amplitude of +-CP light reflected on the left, and transmitted to the right, respectively.

For consideration of propagation in either direction we find it convenient to use a space-fixed (rather than $\hat{\mathbf{k}}$-fixed, where $\hat{\mathbf{k}}$ gives the propagation direction) coordinate system. We resolve any polarization state in terms of the two orthogonal + and − circular polarizations. The latter are space-fixed as well, that is, they are invariant under the change of sign $\hat{\mathbf{k}} \to -\hat{\mathbf{k}}$.

Our most general experiment then has four inputs—incident amplitudes for + CP and −CP on the left (l_+, l_-) and right (r_+ and r_-)—and similarly four outputs. The input and output vectors are then given by

$$\mathbf{i} = \begin{bmatrix} l_+ \\ r_+ \\ l_- \\ r_- \end{bmatrix}^i \quad \text{and} \quad \mathbf{o} = \begin{bmatrix} l_+ \\ r_+ \\ l_- \\ r_- \end{bmatrix}^o . \tag{1}$$

In linear response we have $\mathbf{o} = \underline{S}\mathbf{i}$, where the scattering matrix $\underline{S}$ is, in our basis,

$$\underline{S} = \begin{bmatrix} \begin{bmatrix} R^l_{++} & T^r_{++} \\ T^l_{++} & R^r_{++} \end{bmatrix} & \begin{bmatrix} R^l_{+-} & T^r_{+-} \\ T^l_{+-} & R^r_{+-} \end{bmatrix} \\ \begin{bmatrix} R^l_{-+} & T^r_{-+} \\ T^l_{-+} & R^r_{-+} \end{bmatrix} & \begin{bmatrix} R^l_{--} & T^r_{--} \\ T^l_{--} & R^r_{--} \end{bmatrix} \end{bmatrix} . \tag{2}$$

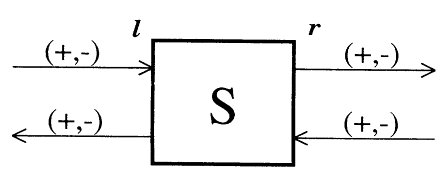

FIG. 2. Schematic of our idealized, general light-scattering experiment. Monochromatic, fully-polarized light, resolved in the circular basis (+ CP or − CP) is normally incident from the left (l) or right (r). Reflected and transmitted light is resolved in the same basis. $\underline{S}$ is the full scattering matrix giving the outputs as a linear function of the inputs.

$\underline{S}$ characterizes the sample: Invariance of the latter under a given symmetry operation $\hat{\mathbf{O}}$ implies the invariance of $\underline{S}$ under the same. Hence we examine, in our basis, the effects of several symmetry operations on the matrix $\underline{S}$.

A. Space inversion $\mathcal{P}$

Space inversion ($\mathbf{r}\to-\mathbf{r}$) exchanges left and right surfaces of the sample and changes the sign of the wave vector $\mathbf{k}$ of the light, while leaving the CP unchanged ($\mathcal{P}$ changes the *handedness* but not the absolute circular rotation). Thus,

$$\hat{\underline{\mathcal{P}}}\begin{bmatrix} l_+ \\ r_+ \\ l_- \\ r_- \end{bmatrix}^{i,o} = \begin{bmatrix} r_+ \\ l_+ \\ r_- \\ l_- \end{bmatrix}^{i,o} \tag{3}$$

and so

$$\hat{\underline{\mathcal{P}}}=\begin{bmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{bmatrix} = \begin{bmatrix} \sigma_x & \underline{0}_{2\times2} \\ \underline{0}_{2\times2} & \sigma_x \end{bmatrix} = \hat{\underline{\mathcal{P}}}^{-1} , \tag{4}$$

where σ_x is the usual 2×2 Pauli matrix $\left(\begin{smallmatrix} 0 & 1 \\ 1 & 0 \end{smallmatrix}\right)$. The scattering matrix for the inverted sample is then

$$\hat{\underline{\mathcal{P}}}\,\underline{S}\,\hat{\underline{\mathcal{P}}}^{-1} = \begin{bmatrix} \begin{bmatrix} R^r_{++} & T^l_{++} \\ T^r_{++} & R^l_{++} \end{bmatrix} & \begin{bmatrix} R^r_{+-} & T^l_{+-} \\ T^r_{+-} & R^l_{+-} \end{bmatrix} \\ \begin{bmatrix} R^r_{-+} & T^l_{-+} \\ T^r_{-+} & R^l_{-+} \end{bmatrix} & \begin{bmatrix} R^r_{--} & T^l_{--} \\ T^r_{--} & R^l_{--} \end{bmatrix} \end{bmatrix} . \tag{5}$$

B. Time reversal $\mathcal{T}$

In considering the effects of time reversal $\mathcal{T}$ on a scattering system one may invoke the known symmetry properties of linear-response coefficients in Cartesian coordinates;[20] alternatively, one may invoke the principle of reciprocity, which has been stated in various forms over the last century.[16,17] Here we choose the former approach. For an application of the principle of reciprocity to the components of the scattering matrix $\underline{S}$ see Ref. 15 and the Appendix.

In linear response, and in Cartesian (α,β) coordinates, the electric field at $\mathbf{r}'$ arising from an input field at $\mathbf{r}$ is

$$E_\alpha(\mathbf{r}')=\sum_\beta \chi_{\alpha\beta}(\mathbf{r},\mathbf{r}')E_\beta(\mathbf{r}) . \tag{6}$$

We choose $\hat{\mathbf{k}}\|\hat{\mathbf{z}}$ from now on, so that (α,β) range over (x,y). χ is then a 2×2 matrix that transforms under time reversal as[20]

$$\bar{\chi}\equiv\chi(\mathbf{r}',\mathbf{r},-\Phi)=\chi^T(\mathbf{r},\mathbf{r}',\Phi) . \tag{7}$$

Here Φ represents any quantities odd under $\mathcal{T}$, such as an external magnetic field, or, in our case of interest, a time-odd order parameter of the sample.

We now temporarily abstract the 2×2 reflection and transmission matrices for right and left inputs ($\underline{R}^r,\underline{R}^l$, etc.) from our 4×4 scattering matrix $\underline{S}$. Letting $\mathbf{r}$ and $\mathbf{r}'$ coincide on one side of the sample, we then get

$$\bar{\underline{R}}^{l,r}_{\text{car}}=(\underline{R}^{l,r}_{\text{car}})^T . \tag{8}$$

For $\mathbf{r}$ and $\mathbf{r}'$ on opposite sides we get the transformation law for the transmission matrices:

$$\bar{\underline{T}}^{l,r}_{\text{car}}=(\underline{T}^{r,l}_{\text{car}})^T . \tag{8$'$}$$

We can express these in our circular basis using $\underline{X}_{\text{circ}}=\underline{C}\,\underline{X}_{\text{car}}\underline{C}^{-1}$ where

$$\underline{C}=\frac{1}{\sqrt{2}}\begin{bmatrix} 1 & -i \\ 1 & i \end{bmatrix} .$$

Changing to the circular basis, and reassembling everything in our matrix S, we get

$$\bar{\underline{S}}=\underline{S}(-\Phi)=\begin{bmatrix} \begin{bmatrix} R^l_{--} & T^l_{--} \\ T^r_{--} & R^r_{--} \end{bmatrix} & \begin{bmatrix} R^l_{+-} & T^l_{+-} \\ T^r_{+-} & R^r_{+-} \end{bmatrix} \\ \begin{bmatrix} R^l_{-+} & T^l_{-+} \\ T^r_{-+} & R^r_{-+} \end{bmatrix} & \begin{bmatrix} R^l_{++} & T^l_{++} \\ T^r_{++} & R^r_{++} \end{bmatrix} \end{bmatrix} , \tag{9}$$

or more simply

$$\bar{\underline{S}}=\hat{\underline{\mathcal{J}}}\,\underline{S}^T\hat{\underline{\mathcal{J}}}^{-1} , \tag{10}$$

where

$$\hat{\underline{\mathcal{J}}}=\begin{bmatrix} \underline{0}_{2\times2} & \underline{1}_2 \\ \underline{1}_2 & \underline{0}_{2\times2} \end{bmatrix}=\hat{\underline{\mathcal{J}}}^{-1} . \tag{11}$$

C. Rotations about the propagation direction

The effect of a rotation by an angle θ about $\hat{\mathbf{k}}$ on any polarization state, expressed in our (two-dimensional) circular basis, is

$$\begin{bmatrix} e^{i\theta} & 0 \\ 0 & e^{-i\theta} \end{bmatrix}$$

so that the rotation matrix in our 4×4 notation is

$$\hat{\underline{\mathcal{R}}}(\theta)=\begin{bmatrix} e^{i\theta}1_2 & 0_{2\times2} \\ 0_{2\times2} & e^{-i\theta}1_2 \end{bmatrix} . \tag{12}$$

D. Reflections

Again the xy plane is perpendicular to $\hat{\mathbf{k}}$. Define $\hat{\mathcal{Y}}$ to be the operator for reflection about the y axis ($x\to-x$). Then

$$\hat{\underline{\mathcal{Y}}}_{\text{car}}=\begin{bmatrix} -1 & 0 \\ 0 & 1 \end{bmatrix}$$

and

$$\hat{\underline{\mathcal{Y}}}_{\text{circ}}=-\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} .$$

In our 4×4 circular basis

$$\hat{\underline{Y}} = -\begin{bmatrix} \underline{0}_{2\times 2} & 1_2 \\ 1_2 & \underline{0}_{2\times 2} \end{bmatrix} = -\hat{\underline{J}} \quad (13)$$

and similarly $\hat{\underline{X}}(y \to -y) = +\hat{\underline{J}}$.

E. Combined symmetries

If a sample is invariant under combined $\mathcal{PT}$, then

$$\underline{S} = \begin{bmatrix} \begin{bmatrix} R^l_{++} & T^r_{++} \\ T^l_{++} & R^r_{++} \end{bmatrix} & \begin{bmatrix} R^l_{+-} & T^r_{+-} \\ T^l_{+-} & R^r_{+-} \end{bmatrix} \\ \begin{bmatrix} R^l_{-+} & T^r_{-+} \\ T^l_{-+} & R^r_{-+} \end{bmatrix} & \begin{bmatrix} R^l_{--} & T^r_{--} \\ T^l_{--} & R^r_{--} \end{bmatrix} \end{bmatrix}$$

$$= \hat{\underline{P}}\hat{\underline{J}}\,\underline{S}^T\hat{\underline{J}}\hat{\underline{P}} = \begin{bmatrix} \begin{bmatrix} R^r_{--} & T^r_{--} \\ T^l_{--} & R^l_{--} \end{bmatrix} & \begin{bmatrix} R^r_{+-} & T^r_{+-} \\ T^l_{+-} & R^l_{+-} \end{bmatrix} \\ \begin{bmatrix} R^r_{-+} & T^r_{-+} \\ T^l_{-+} & R^l_{-+} \end{bmatrix} & \begin{bmatrix} R^r_{++} & T^r_{++} \\ T^l_{++} & R^l_{++} \end{bmatrix} \end{bmatrix} . \quad (14)$$

In our model, the sample is composed of many planes, each of which breaks $\mathcal{T}$ symmetry. For orthorhombic or tetragonal crystals (appropriate to the HTSC) the sample is invariant under $\hat{\underline{Y}}$ in the absence of the broken $\mathcal{T}$ symmetry. Given the latter, however, each plane (and thus the entire sample) is invariant under combined $\hat{\underline{Y}}\mathcal{T}$ (which is called "PT" in the anyon literature[9]). Since, however, $\hat{\underline{J}} = \hat{\underline{J}}^{-1}$, $\hat{\underline{Y}}\mathcal{T}$ invariance gives simply

$$\underline{S}^T = \underline{S} \ . \quad (15)$$

The other symmetry relevant to orthorhombic or tetragonal crystals is rotational symmetry. Orthorhombic rotational symmetry gives no constraint on $\underline{S}$ [since $\hat{\underline{R}}(\pi) = -\underline{1}_4$]; however, tetragonal (fourfold) rotational symmetry requires the off-diagonal blocks of $\underline{S}$ to be zero:

$$\underline{S} = \begin{bmatrix} \underline{S}_{++} & \underline{0}_{2\times 2} \\ \underline{0}_{2\times 2} & \underline{S}_{--} \end{bmatrix} . \quad (16)$$

Finally, we summarize the relations obtained for perfect crystals of two magnetic point groups:[21] $m'm'm'$, which is in our notation $[\mathcal{PT} + \hat{\underline{Y}}\mathcal{T} + \hat{\underline{R}}(\pi)]$ (orthorhombic); and $4/m'm'm'$, or $[\mathcal{PT} + \hat{\underline{Y}}\mathcal{T} + \hat{\underline{R}}(\pi/2)]$ (tetragonal). For $m'm'm'$ we get

$$\begin{aligned} &T^l_{++} = T^r_{++} = T^l_{--} = T^l_{--} \equiv T_d \ , \\ &R^l_{++} = R^r_{--} \ , \\ &R^r_{++} = R^l_{--} \ , \\ &R^l_{+-} = R^l_{-+} = R^r_{-+} = R^r_{+-} \equiv R_{od} \ , \\ &T^r_{+-} = T^l_{-+} \ , \\ &T^l_{+-} = T^r_{-+} \ . \end{aligned} \quad (17)$$

For the tetragonal case we simply set all the off-diagonal $[(+-)$ and $(-+)]$ elements to zero in Eq. (17).

F. Application to optical rotation

The above results may now be applied to known experiments on optical rotation in the HTSC. In each case we wish to test the compatibility of the $\mathcal{PT}$ invariant model with experiment, in terms of the constraints derived purely from symmetry.

Early experiments by Lyons *et al.*[3] detected a signal proportional to

$$(|R_{++}|^2 - |R_{--}|^2) + (|R_{+-}|^2 - |R_{-+}|^2) \equiv S_{L1} \ .$$

As shown by Halperin,[10] and confirmed by our Eqs. (2) and (9), a nonzero value of $(|R_{++}|^2 - |R_{--}|^2)$ is evidence for broken $\mathcal{T}$ symmetry of the sample; however S_{L1} contains other terms that complicate the interpretation of the results. Hence, later experiments by the same group[4] used a modified optical apparatus giving a signal proportional to $(|R_{++}|^2 - |R_{--}|^2) \equiv S_{L2}$. The positive results from these experiments appear to give unambiguous evidence of broken $\mathcal{T}$ symmetry in the HTSC. We note from Eq. (14) that $\mathcal{PT}$ symmetry also allows $S_{L2} \neq 0$, as does [from Eq. (17)] an orthorhombic 1:2:3 crystal with AFM order of the planes.

Lyons *et al.* have also measured $(|R_{++}|^2 - |R_{--}|^2)$ for a single sample of 2:1:4. As noted by Dzyaloshinskii,[13] this material, with one plane per unit cell, does not break $\mathcal{T}$ or $\mathcal{P}$ (neglecting surface effects) if the planes order antiferromagnetically. It is thus interesting that a null result was found[4] for S_{L2} for this material. Unfortunately, to date only a single sample has been examined. This result, if replicated, coupled with the positive results for 1:2:3, would be strong evidence in favor of the $\mathcal{PT}$-invariance hypothesis.

Experiments by Spielman *et al.*,[5,6] using the Sagnac interferometer at Stanford University measured $\arg(T^l_{++}/T^r_{--}) \equiv S_{S1}$. From our Eqs. (2) and (9) this quantity is zero if $\mathcal{T}$ symmetry is unbroken in the sample. $\mathcal{PT}$ symmetry by itself does not compel $S_{S1} = 0$ (even though, as pointed out by Dzyaloshinskii,[13] $\mathcal{PT}$ symmetry is sufficient to give $T^i_{++} = T^i_{--}$). However, the additional symmetries $(m'm'm')$ embodied in Eq. (17) suffice to ensure that $S_{S1} = 0$. Hence, at the present level of analysis (neglecting questions of defects, polycrystallinity, etc.), the transmission results of Spielman *et al.* are consistent with the $\mathcal{PT}$-invariance hypothesis.

More recently,[7] Spielman *et al.* have modified their apparatus to allow for interferometric reflection measurements. Their signal is then $\arg(R_{++}/R_{--}) \equiv S_{S2}$. Since the $\mathcal{PT}$-invariant model, as well as any model giving broken $\mathcal{T}$ symmetry alone, predicts $R^i_{++} \neq R^i_{--}$ $(i = r, l)$, the null result of this experiment disagrees with the $\mathcal{PT}$-invariance hypothesis in the weak sense that an effect which is allowed is not seen. This result is also in nearly direct contradiction with the finding of Lyons *et al.*[4] mentioned above.

Weber *et al.*[8] reported optical rotation in both reflection and transmission experiments. For the former, it is not clear from their analysis how to translate their measured signal into our notation. We can say that their signal ($\zeta_A-\zeta_B$ in their notation) is composed of a term proportional to $\mathrm{Re}(R_{++}-R_{--})$ and a term proportional to $\mathrm{Im}(R_{++}-R_{--})$ (these sensitive to broken $\mathcal{T}$), and a part involving R_{+-} and R_{-+}. Weber *et al.* emphasize the former terms as being responsible for their observed signal, based on the dependence of the sign of $\zeta_A-\zeta_B$ on applied magnetic fields in field-cooling experiments. As we have noted already, $R_{++}\neq R_{--}$ is allowed by the $\mathcal{PT}$-invariant model.

For the transmission experiments the input polarization was aligned with a principal axis of the (2:2:1:2) crystal; this procedure was possible, since the twin size exceeded the laser spot size for these samples. The rotation angle of the transmitted light was then measured. To make contact with our notation we let $\hat{\mathbf{x}}$ be the principal axis and assume incidence on the left; the azimuth of the output is then

$$-\frac{1}{2}\arg\left[\frac{T^l_{++}+T^l_{+-}}{T^l_{-+}+T^l_{--}}\right]\equiv S_{W2}\ . \tag{18}$$

This quantity is allowed to be nonzero by $\mathcal{PT}$ symmetry alone. Adding the orthorhombic and $\underline{\hat{Y}}\mathcal{T}$ symmetries gives

$$S_{W2}=-\frac{1}{2}\arg\left[\frac{T_d+T^l_{+-}}{T_d+T^l_{-+}}\right]\ , \tag{19}$$

while increasing the symmetry to tetragonal gives $S_{W2}=-\frac{1}{2}\arg(T_d/T_d)=0$.

The $\mathcal{PT}$-invariant model is thus consistent with the results of the Weber transmission experiment only if we assume that the samples used were not rotationally invariant about the c axis, i.e., not tetragonal. This assumption is, however, reasonable; although 2:2:1:2 is nominally tetragonal[13] there is, in fact, ample evidence for significant structural[8,22] and optical[6,23] anisotropy in the *ab* plane.

Later experiments by Weber *et al.*[8] measured "average ellipticity" $\varepsilon_{\mathrm{av}}\equiv S_{W3}$ in transmission through 1:2:3 films, for linearly polarized input. Here average ellipticity means the average for two orthogonal input polarizations, the idea being to cancel the ellipticity arising from linear birefringence. The measured quantity $\varepsilon_{\mathrm{av}}$ (thus defined) is in fact not equivalent[24] to

$$\varepsilon_{\mathrm{CD}}\equiv\varepsilon|_{T_{+-}=T_{-+}=0}$$
$$=[(|T_{++}|-|T_{--}|)/(|T_{++}|+|T_{--}|)]\ .$$

The $\mathcal{PT}$ model gives $\varepsilon_{\mathrm{CD}}=0$ but $\varepsilon_{\mathrm{av}}\neq 0$.

We thus find that the $\mathcal{PT}$-invariant model, i.e., broken $\mathcal{T}$ symmetry in the planes, ordered antiferromagnetically, is consistent with all but one of the above results on optical rotation in the HTSC. The result which is not predicted by our symmetry analysis is the null result of Speilman *et al.* for $S_{S2}=\arg(R_{++}/R_{--})$. This result, being null, is not forbidden by the $\mathcal{PT}$-invariance hypothesis; but it does not support it. Below (Sec. IV) we will discuss in more detail the possible resolution of these results. First, in the next section, we present a phenomenological realization of the $\mathcal{PT}$-invariant model in terms of a slab with nonzero magnetoelectric coefficients.

III. MAGNETOELECTRIC MODEL

A. Symmetry properties

As shown by Dzyaloshinskii,[13,19] a consequence of $\mathcal{PT}$ symmetry is the appearance of nonzero magnetoelectric coefficients in the constitutive relations for the material, which take the form

$$\begin{bmatrix}\mathbf{D}\\ \mathbf{B}\end{bmatrix}=\begin{bmatrix}\underline{\hat{\epsilon}} & \underline{\hat{\alpha}}^{EH}\\ \underline{\hat{\alpha}}^{HE} & \underline{\hat{\mu}}\end{bmatrix}\begin{bmatrix}\mathbf{E}\\ \mathbf{H}\end{bmatrix}\ . \tag{20}$$

Following a notation similar to that of Berreman,[25] we can write the above more concisely as

$$\mathbf{C}=\underline{M}\mathbf{G}\ . \tag{21}$$

It is useful to decompose the (4×4) linear-response tensor $\underline{M}$ as

$$\underline{M}=\begin{bmatrix}\underline{\hat{\epsilon}} & \underline{0}_{2\times 2}\\ \underline{0}_{2\times 2} & \underline{\hat{\mu}}\end{bmatrix}+\begin{bmatrix}\underline{0}_{2\times 2} & \underline{\hat{\alpha}}^{EH}\\ \underline{\hat{\alpha}}^{HE} & \underline{0}_{2\times 2}\end{bmatrix}\equiv\underline{\hat{\kappa}}+\underline{\hat{\alpha}}\ ; \tag{22}$$

that is, the off-diagonal blocks are confined to $\underline{\hat{\alpha}}$. With this notation in place we can briefly discuss the symmetry properties of the tensor $\hat{\alpha}$.

Onsager reciprocity relations give $\underline{\hat{\alpha}}^{EH}(\Phi)=-(\underline{\hat{\alpha}}^{HE})^T(-\Phi)$ where Φ is again a $\mathcal{T}$-symmetry-breaking order parameter. Hence we find that the $\mathcal{T}$-invariant part of $\underline{\hat{\alpha}}$ must be antisymmetric, or, conversely, the symmetric part of $\underline{\hat{\alpha}}$ gives broken $\mathcal{T}$ symmetry.

By the same reasoning[26] as that applied to the tensor $\hat{\kappa}$, we find that the Hermitian part of $\hat{\alpha}$ is nondissipative, and the antihermitian part dissipative. Hence we find that the form of $\hat{\alpha}$ appropriate to the $\mathcal{PT}$-invariant model we wish to investigate is a symmetric tensor whose real part is nondissipative and whose imaginary part gives dissipation.

It is interesting to note that the magnetoelectric tensor $\hat{\alpha}$ has also been associated with optical activity,[25,27–29] a phenomenon that is $\mathcal{T}$ invariant. We see from the above that a description of optical activity in terms of $\hat{\alpha}$ can only be correct if $\hat{\alpha}$ is antisymmetric, and hence, for nondissipative processes, purely imaginary. Problems arise[30] (e.g. $R_{++}\neq R_{--}$) when this constraint is not adhered to.

We return to the $\mathcal{PT}$-invariant model, and again restrict our considerations to propagation along the c ($\|\hat{\mathbf{z}}$) axis. This allows us to neglect the z components of all the tensors in M, rendering it a 4×4 matrix whose blocks are 2×2. For the magnetic point symmetry $m'm'm'$ ($\mathcal{PT}$ + orthorhombic) the form of $\underline{\hat{\alpha}}^{EH}$ must be[21] $\mathrm{diag}(\alpha_{11},\alpha_{22})$, while for the tetragonal case we must have $\alpha_{11}=\alpha_{22}$.

We note that Dzyaloshinskii[13] obtained a symmetric $\hat{\alpha}$ from the many-AFM-plane model of the HTSC. Dzyaloshinskii found that $\hat{\alpha}\propto i\epsilon^a_{xy}$, where ϵ^a_{xy} is the antisymmetric (and hence $\mathcal{T}$-symmetry-breaking) part of the two-dimensional dielectric tensor for each plane (alternating in sign from one plane to the next). This quantity is also rotationally invariant; hence we find that, for this origin for $\hat{\alpha}$, $\alpha_{11}=\alpha_{22}$ regardless of crystal symmetry. Thus, finally, the version of $\hat{\alpha}$ that we wish to consider in the following is the simplest possible: $\underline{\hat{\alpha}}^{EH}=\underline{\hat{\alpha}}^{HE}=\text{diag}(\alpha,\alpha)$.

B. Eigenmode of propagation

We solve the problem of propagation in a magnetoelectric slab in the standard way. We first find the eigenmodes of propagation[31] for an infinite medium; these modes plus the standard boundary conditions then give us the complete solution for reflected and transmitted waves from the slab.

The eigenmodes are conveniently found using the 4×4 method of Berreman.[25] We take $\underline{\hat{\epsilon}}=\text{diag}(\epsilon_1,\epsilon_2)$ ($x=1;y=2$) and $\underline{\hat{\mu}}=\underline{1}_2$. Then, keeping terms to first order in α, we find

$$k_{1,2}=\pm\left[\frac{\omega}{c}\right]\sqrt{\epsilon_1}\,,\qquad k_{3,4}=\pm\left[\frac{\omega}{c}\right]\sqrt{\epsilon_2}\,, \tag{23}$$

for the wave vectors of the eigenmodes, i.e., the same as the unperturbed values to $O(\alpha^2)$. We write the eigenvectors in the form

$$\Psi_i=\begin{bmatrix}E_x\\E_y\\H_x\\H_y\end{bmatrix}, \tag{24}$$

where **E**, **H** are the electromagnetic fields and the time dependence is implicit. The eigenvectors are then, to $O(\alpha^2)$,

$$\Psi_{1,2}=\begin{bmatrix}1\\0\\-\alpha\\\pm n_1\end{bmatrix};\quad \Psi_{3,4}=\begin{bmatrix}0\\1\\\mp n_2\\-\alpha\end{bmatrix}, \tag{25}$$

where $n_{1,2}=\sqrt{\epsilon_{1,2}}$.

We note that these eigenvectors represent light that is linearly polarized along the principal axes of $\hat{\epsilon}$. The particular feature due to a nonzero magnetoelectric coefficient α is that $\mathbf{E}\cdot\mathbf{H}\neq 0$; this feature will be shown to give a rotation at the incident boundary ($\propto\alpha$) such that the reflected wave, and the transmitted wave *internal* to the material, will not lie on a principal axis even when the incident polarization does. Even for the tetragonal case ($\epsilon_1=\epsilon_2$) both the reflected and the internally transmitted waves are rotated with respect to the input. From this point of view, the surprise is then that, for the tetragonal case, the transmitted wave emerges *unrotated* on the far side—as required by our symmetry analysis.

C. The orthorhombic slab

For simplicity we let the incident wave be x-polarized light, assumed incident from the left (Fig. 3). In the notation of Eq. (24), we write the incident wave, the reflected wave, and the transmitted (at the far side of the slab, of thickness d) wave as

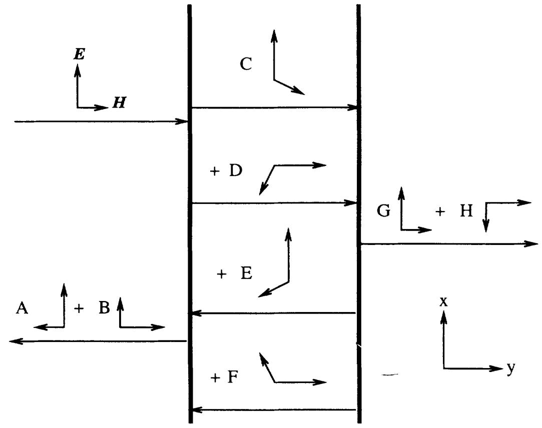

FIG. 3. Wave propagation through a magnetoelectric slab (bounded by the heavy vertical lines). Incident light (upper left) is fully $\hat{x}$ polarized. The xy plane, in which lie the **E** and **H** vectors of the light, and which is normal to the propagation direction, is rotated into the plane of the paper for the purpose of clearly showing **E** and **H** (see coordinate axes for **E** and **H** vectors at lower right); also, **H** is drawn shorter than **E** for clarity. The reflected and transmitted waves are resolved into $\hat{x}$ and $\hat{y}$ components as shown. The complex scalar amplitudes for each eigenmode (internal or external) are denoted $A,B,\ldots,E,\ldots,H$; these should not be confused with the electromagnetic field vectors **E** and **H**. The four eigenmodes internal to the slab are characterized by $\mathbf{E}\cdot\mathbf{H}\neq 0$, due to the nonzero magnetoelectric coefficients of the slab. This gives rise in general to a rotation in transmission ($H\neq 0$) and in reflection ($B\neq 0$) even when $\hat{\mathbf{x}}$ is a principal axis of the dielectric tensor (as shown here).

$$\iota=\begin{bmatrix}1\\0\\0\\1\end{bmatrix}e^{-ikz},\qquad \mathbf{r}=\begin{bmatrix}A\\B\\B\\-A\end{bmatrix}e^{-ikz},\qquad \mathbf{t}=\begin{bmatrix}G\\H\\-H\\G\end{bmatrix}e^{ik(z-d)}. \tag{26}$$

The internal waves are written in terms of the above eigenmodes:

$$C\Psi_1 e^{ik_1 z}+E\Psi_2 e^{-ik_1 z}+D\Psi_3 e^{ik_2 z}+F\Psi_4 e^{-ik_2 z}.$$

The eight unknowns $A-H$ are then found from standard boundary conditions at the two surfaces.

We define $p_1\equiv e^{ik_1 d}$, $p_2\equiv e^{ik_2 d}$, and $A\equiv N_A/\mathcal{D}$, $B\equiv N_B/\mathcal{D}$, etc., where $\mathcal{D}$ is the common denominator for the eight unknowns. We also define

$$\begin{aligned} Q&=(n_2+1)(n_1+1)\ ,\\ R&=(n_2-1)(n_1+1)\ ,\\ S&=(n_2+1)(n_1-1)\ ,\\ T&=(n_2-1)(n_1-1)\ . \end{aligned} \tag{27}$$

Then the waves in the slab are given by

$$\begin{aligned} N_A&=(p_1-p_1^*)[p_2RT-p_2QS]\ ,\\ N_B&=2\alpha[p_1^*p_2^*Q+p_1^*p_2R+p_1p_2^*S+p_1p_2T-4n_1n_2]\ ,\\ N_C&=2p_1^*p_2(n_2-1)R-2p_1^*p_2^*(n_2+1)Q\ ,\\ N_D&=2\alpha[2n_1(1-n_2)+p_1^*p_2^*Q+p_1p_2^*S]\ ,\\ N_E&=2p_1p_2(n_2-1)T-2p_1p_2^*(n_2+1)S\ ,\\ N_F&=2\alpha[p_1^*p_2R+p_1p_2T-2n_1(n_2+1)]\ ,\\ N_G&=4n_1[p_2(n_2-1)^2-p_2^*(n_2+1)^2]\ ,\\ N_H&=4\alpha[p_1^*n_2(n_1+1)+p_1n_2(n_1-1)\\ &\qquad -p_2n_1(n_2-1)-p_2^*n_1(n_2+1)]\ ,\\ \mathcal{D}&=p_1p_2^*S^2-p_1p_2T^2-p_1^*p_2^*Q^2+p_1^*p_2R^2\ , \end{aligned} \tag{28}$$

plus terms of $O(\alpha^2)$.

Since B gives the E_y component of the reflected wave, and H that of the transmitted wave, we see that there is a rotation $\propto\alpha$ in both reflection and transmission, for input polarized along a principal axis. The solutions for y-polarized input (denoted $A', B', \ldots, H'$) may be obtained from the above by letting

$$\begin{pmatrix} A'\\ B'\\ C'\\ D'\\ E'\\ F'\\ G'\\ H' \end{pmatrix} = \begin{pmatrix} B\\ A\\ D\\ C\\ F\\ E\\ H\\ G \end{pmatrix}_{\left(\substack{\alpha\to-\alpha\\ 1\leftrightarrow 2}\right)} . \tag{29}$$

To make contact with the notation of Sec. II, we note that the reflection and transmission matrices, for left-incident light and in Cartesian coordinates, are

$$\begin{aligned} \hat{\underline{R}}^{\,l}_{\rm car}&=\begin{bmatrix} A & A'\\ B & B' \end{bmatrix}\ ,\\ \hat{\underline{T}}^{\,l}_{\rm car}&=\begin{bmatrix} G & G'\\ H & H' \end{bmatrix}\ . \end{aligned} \tag{30}$$

The mapping (29) gives $A'=-B$ and $B'=\tilde{A}$, where $\tilde{X}\equiv X(1\leftrightarrow 2)$. With this notation, and changing to the circular polarization basis of Sec. II, we get

$$\begin{aligned} \hat{\underline{R}}^{\,l}_{\rm circ}&=\begin{bmatrix} \frac{1}{2}(A+\tilde{A})-iB & \frac{1}{2}(A-\tilde{A})\\ \frac{1}{2}(A-\tilde{A}) & \frac{1}{2}(A+\tilde{A})+iB \end{bmatrix}\ ,\\ \hat{\underline{T}}^{\,l}_{\rm circ}&=\begin{bmatrix} \frac{1}{2}(G+\tilde{G}) & \frac{1}{2}(G-\tilde{G})-iH\\ \frac{1}{2}(G-\tilde{G})+iH & \frac{1}{2}(G+\tilde{G}) \end{bmatrix}\ . \end{aligned} \tag{31}$$

The solutions for input from the right are obtained from the above by simply changing the sign of α. Hence

$$\begin{aligned} \hat{\underline{R}}^{\,r}_{\rm circ}&=\hat{\underline{R}}^{\,l}_{\rm circ}(B\to -B)\ ,\\ \hat{\underline{T}}^{\,r}_{\rm circ}&=\hat{\underline{T}}^{\,l}_{\rm circ}(H\to -H)\ . \end{aligned} \tag{32}$$

We see, as a final check on our results, that the above matrices [Eqs. (31) and (32)] satisfy the constraints for the $m'm'm'$ case, given in Eq. (17) above.

D. The tetragonal slab

We obtain the solution for the slab with rotational symmetry about $\hat{\mathbf{z}}$ by setting $1=2$ everywhere in the above. It is readily verified that this gives the expected results: $H=0$ but $B\neq 0$; off-diagonal components of the circular matrices vanish; and $R_{++}\neq R_{--}$ for either side, while (as in the orthorhombic case) $T_{++}=T_{--}$. Hence, again, this case gives rotation in reflection but none in transmission.

IV. DISCUSSION

We find that our analysis of the $\mathcal{PT}$-invariant model for the HTSC—that is, a model in which $\mathcal{T}$ symmetry is broken in each plane, and the planes order antiferromagnetically—offers some promise of reconciling the many confusing experimental ellipsometric results. In fact, it appears that there are only two likely candidate hypotheses that might be compatible with all the experimental results. We would state these two hypotheses as follows: (i) The $\mathcal{PT}$-invariant model is correct; $\mathcal{T}$ symmetry is spontaneously broken in the normal state of the HTSC, and the planes order antiferromagnetically. (ii) $\mathcal{T}$ symmetry is not spontaneously broken in the doped phase of the HTSC.

There is also, of course, the possibility that $\mathcal{T}$ symmetry is broken in each plane, with the planes ordered ferromagnetically—along with other conceivable schemes for ordering the planes. There are, however, theoretical arguments[11,12] against ferromagnetic (FM) order of the planes. Furthermore, we believe that FM order of the planes should give macroscopic signatures of the broken $\mathcal{T}$ symmetry, which would be detected in other experiments, such as μSR experiments[2] seeking internal magnetic fields. For these reasons, we view the FM hypothesis as less likely than either (i) or (ii) above, and so we concentrate on the latter two hypotheses in the following.

Neither (i) nor (ii) can explain all the available results without some recourse to technical questions other than symmetry. We would like to briefly discuss some of these questions below. We will also suggest two possible experimental tests, based on our symmetry analysis, of the $\mathcal{PT}$-invariance hypothesis (i).

As we have noted in Sec. II, the $\mathcal{PT}$-invariance hypothesis is compatible with all known ellipsometric results save one. The challenge to (i) is the null result of Spielman *et al.*[7] for $\arg(R_{++}/R_{--})$. The $\mathcal{PT}$-invariant model predicts that, for the materials examined in this experiment (1:2:3 and 2:2:1:2, each with two planes per unit cell), $R_{++}\neq R_{--}$ in general. It is of interest that, in one case involving sample exchange, Spielman *et al.* found $\arg(R_{++}/R_{--})=0$ for the *same sample* for which Lyons *et al.*[4] found $|R_{++}|^2-|R_{--}|^2\neq 0$. A possible explanation for this discrepancy involves the finite size of the broken-$\mathcal{T}$-symmetry domains in each plane. There is evidence from experiments with ferromagnetic garnets[4,7] that the apparatus used by Spielman *et al.*[5–7] is less efficient in resolving multiple domains, if the latter are on the order of a few micrometers in extent. (We note that, if we were to reject all results of Spielman *et al.* for this reason, *and* disregard the difference seen by Lyons *et al.* in a single sample of 2:1:4, then the FM hypothesis would remain as a viable explanation for the remaining optical results; the objections to this hypothesis, mentioned above, of course remain.)

Hypothesis (ii) requires the rejection of all results implying broken $\mathcal{T}$ symmetry in the HTSC. Since the results of Weber *et al.*[8] are not free of possible contamination from $\mathcal{T}$-invariant artifacts (such as spontaneous lattice distortion with diminishing temperature), these results do not offer as strong a challenge to (ii) as do those of Lyons *et al.* The latter results however directly contradict (ii). That is, assuming these results are correct, $\mathcal{T}$ symmetry *must* be broken in the HTSC.

This contradiction is much stronger than the apparent disagreement between the hypothesis (i) and the reflection results of Spielman *et al.*: The results of Lyons *et al.* are *strictly forbidden* by the null hypothesis (ii), while the reflection results of Spielman *et al.* only fail to report an effect allowed by (i). In other words, if we assume that all the experimental results are correct as reported, then simple logic forces the rejection of (ii), leaving us with (i)—the $\mathcal{PT}$-invariance hypothesis. It remains possible, of course, that the *premise* of the previous sentence is not valid; we note that, to date, none of the reported results have been replicated.

Two tests of the $\mathcal{PT}$-invariance hypothesis are suggested by our analysis. The first involves $S_{L2}=(|R_{++}|^2-|R_{--}|^2)$. As pointed out in Ref. 14, this quantity can serve to distinguish ferromagnetic or "simple" broken $\mathcal{T}$ symmetry from the antiferromagnetic $\mathcal{PT}$ symmetry. Define

$$D_1\equiv S^l_{L2}-S^r_{L2}=(|R^l_{++}|^2-|R^l_{--}|^2)-(|R^r_{++}|^2-|R^r_{--}|^2)\ . \tag{33}$$

This quantity is zero for the ferromagnetic case, and equal to $2S^l_{L2}$ for the $\mathcal{PT}$-invariant case. (It is trivially zero for cases in which $\mathcal{T}$ symmetry is unbroken.) Hence the measurement of a nonzero value for D_1 would uniquely and unambiguously indicate $\mathcal{PT}$ symmetry.

A second test involves a transmission measurement like that of Weber *et al.*[8] The quantity S_{W2} defined above (Sec. II) is simply the azimuthal rotation in transmission for principal-axis input. Abbreviating this quantity as $\Theta^{l,r}$, where the superscript indicates the incident surface, define

$$D_2\equiv S^l_{W2}-S^r_{W2}=\Theta^l-\Theta^r\ . \tag{34}$$

This quantity is also zero for a ferromagnet (i.e., for Faraday rotation), and $2\Theta^l$ for the $\mathcal{PT}$-invariant state. D_2 allows a further discrimination: If an observed Θ in a transmission measurement arises from a *misalignment* of the principal axes (arising with decreasing temperature), and hence from simple linear birefringence rather than spontaneously broken $\mathcal{T}$ symmetry, then D_2 will be zero. D_2 does not, however, distinguish rotation due to broken $\mathcal{P}$ and $\mathcal{T}$ symmetries (i.e., the $\mathcal{PT}$-invariant state) from rotation due to spontaneously broken $\mathcal{P}$ symmetry alone, for example, from a spontaneous "twist" in the sample.[8]

Each of these tests requires the availability of nearly perfect samples, such that the AFM order is maintained from one surface to the other. Clearly, the domains must also be sufficiently large to allow their positive identification from either face of the sample. It is not clear whether these requirements can be met with existing crystals. Of the three experimental groups discussed above, Weber *et al.*[8] reports the largest twin size (~ 300 μm); it is commonly assumed that twin size is an upper bound for the size of broken-$\mathcal{T}$-symmetry domains.

V. SUMMARY

Although a number of experiments have been carried out over the past two years, seeking unambiguous evidence of broken time-reversal ($\mathcal{T}$) symmetry in the HTSC, the question remains unresolved. It seems clear that the most promising hypotheses are (i) antiferromagnetic order of broken $\mathcal{T}$ symmetry in the planes, and (ii) no broken $\mathcal{T}$ symmetry in the HTSC. Our analysis shows that (i) is compatible with all the known results, and is supported by all but one. In contrast, (ii) is contradicted by the results of Lyons *et al.*[4] Hence (i) (the "$\mathcal{PT}$-invariance hypothesis") appears stronger than (ii) at this time. However, because of the near contradiction between the reflection experiments of Lyons *et al.*[4] and those of Spielman *et al.*,[7] and the lack of other corroborating experiments to replicate the reported results, one cannot place too much confidence in either (i) or (ii). We hence await further experiments that may resolve the question.

We have suggested two experimental tests for the $\mathcal{PT}$-invariant model. Each requires extremely high-quality crystals; hence it is unclear whether or not these tests will be feasible in the near future. We point out however that one of these proposed experiments—that based on measurement of the quantity D_1 (Sec. IV above)—could serve as a decisive test of the $\mathcal{PT}$-invariance hypothesis. There is therefore, in our view, considerable motivation for seeking to overcome the problems in sample preparation that may stand as obstacles to performing these tests.

Note added. Since this work was submitted, it has been pointed out by Spielman *et al.*[7] and by Shelankov[32] that the presence of a substrate (in thin-film experiments) invalidates our symmetry analysis, since the (film + substrate) does not obey $\mathcal{PT}$ symmetry. A particular consequence of this[32] (which can be verified using the approach of Sec. III) is that a nonreciprocal rotation is expected for the transmission experiments of Spielman *et al.*,[5] i.e., $S_{S1}\propto(n_s-1)\alpha$ where n_s is the substrate dielectric constant and α the magnetoelectric coefficient. Also, Lawrence, Szöke, and Laughlin[33] have repeated the experiment of Lyons *et al.*[3] for 1:2:3 single crystals and thin films, but failed to replicate their positive result. Obviously, these developments are each discouraging for the $\mathcal{PT}$-invariance hypothesis, tending instead to favor the null hypothesis of unbroken $\mathcal{T}$ symmetry in the HTSC.

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APPENDIX: ONSAGER RECIPROCITY AND THE PRINCIPLE OF RECIPROCITY

The principle of reciprocity (POR) was given precise statement in reference to light-scattering experiments by Perrin,[17] although its original statement dates from much earlier.[16] More recent (and concise) statements, and applications, may be found in De Figueiredo and Raab,[17] and in Graham.[17] In this appendix we will use "POR" to denote the statement of Perrin. The POR then relates the intensities of scattered light in an experiment e_1 and in its time-reversed counterpart e_2, analyzed in terms of *linear* polarizations, as follows (see also Fig. 4): Assume that, in experiment e_1, light of intensity I_1 and polarization p_1 is incident on a sample $\underline{S}$ from $\mathbf{r}$, and further that scattered light of intensity I_1' is detected at $\mathbf{r}'$ after passing a polarizer that transmits only p_1'. In experiment e_2, light of intensity I_2 and polarization $p_2 = p_1'$ is incident on the time-reversed sample $\bar{\underline{S}}$ from $\mathbf{r}'$, and scattered light of intensity I_2' is detected at $\mathbf{r}$ after passing a polarizer that transmits $p_2' = p_1$. The POR then states that if $I_2 = I_1$, then $I_2' = I_1'$.

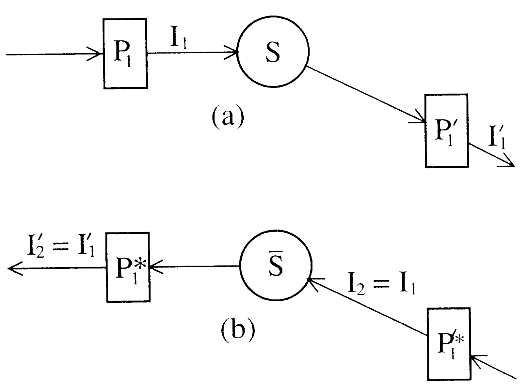

FIG. 4. The principle of reciprocity for general (elliptical) polarizations (denoted POR* in the text). In experiment (a) light of polarization p_1 and intensity I_1, coming from $\mathbf{r}$, is scattered by sample $\underline{S}$ to $\mathbf{r}'$. After passing through polarizer $\hat{P}_1'$ its intensity at $\mathbf{r}'$ is I_1'. In the time-reversed experiment (b), light of polarization $p_2 = p_1'^*$ is incident from $\mathbf{r}'$ with intensity I_2. After scattering from the time-reversed sample $\bar{\underline{S}}$, and passing through the polarizer $\hat{P}_1^*$, light of intensity I_2' is detected at $\mathbf{r}$. The POR* then states that if $I_2 = I_1$, then $I_2' = I_1'$.

In the above (and everywhere in this appendix), $\underline{S} \equiv \underline{S}(\mathbf{r}, \mathbf{r}', \Phi)$ is a 2×2 scattering matrix expressed in the chosen polarization basis; Φ again represents any $\mathcal{T}$-odd aspects of the scattering system. The time-reversed system is then described by $\bar{\underline{S}} \equiv \underline{S}(\mathbf{r}', \mathbf{r}, -\Phi)$. The POR appears to constrain only the magnitudes of the components of $\bar{\underline{S}}$; but in fact, as shown by Shelankov and Pikus[15] (SP), it can be used to constrain the phases as well.

The Onsager reciprocity relations (ORR) used in Sec. II give the magnitude and phase of $\bar{\underline{S}}$ in terms of $\underline{S}$. For a Cartesian basis, the ORR state that $\bar{\underline{S}} = \underline{S}^T$. SP used a "weak" statement of the POR, wherein the emitted and detected intensities are unpolarized; they showed that

$$\text{weak POR} \Longrightarrow \text{ORR} . \tag{A1}$$

(It is easy to show the converse ORR $\Longrightarrow$ weak POR.) In the following we state the POR in a form slightly more general than that of Perrin (we call this the POR*) and then show that

$$\text{POR}^* \Longrightarrow \text{ORR} . \tag{A2}$$

The POR* consists of the POR with the following modifications.[34] Let p_1 and p_1' be *arbitrary* (complex) polarizations; then, in the (time-reversed) experiment e_2 [part (b) of Fig. 4], the polarizations p_2 and p_2' must be the time-reversed counterparts of p_1' and p_1, respectively, i.e., $p_2 = (p_1')^*$ and $p_2' = p_1^*$.

Now we establish some notation. We retain a Cartesian basis, and write p_1 as $\binom{x}{y}$. Then the transfer function for the polarizer $\hat{P}_1$ giving p_1 is

$$\hat{\underline{P}}_1 = \frac{1}{|x|^2 + |y|^2} \begin{pmatrix} x \\ y \end{pmatrix} (x^* \;\; y^*) . \tag{A3}$$

For convenience we set $I_1 = |x|^2 + |y|^2 = 1$. The light emerging at $\mathbf{r}'$, after passing through $\hat{P}_1'$, is

$$\begin{pmatrix} x' \\ y' \end{pmatrix} = \hat{\underline{P}}_1' \underline{S} \begin{pmatrix} x \\ y \end{pmatrix} . \tag{A4}$$

I_1' is then

$$\begin{aligned} I_1' &= (x'^* \;\; y'^*) \begin{pmatrix} x' \\ y' \end{pmatrix} \\ &= (x^* \;\; y^*) \underline{S}^\dagger (\hat{\underline{P}}_1')^\dagger \begin{pmatrix} x' \\ y' \end{pmatrix} \\ &= (x^* \;\; y^*) \underline{S}^\dagger \begin{pmatrix} x' \\ y' \end{pmatrix} . \end{aligned} \tag{A5}$$

For the time-reversed experiment e_2 we note that

$$\bar{\underline{P}}_1=\hat{\underline{P}}_1^*=\hat{\underline{P}}_1^T \tag{A6}$$

and similarly for $\hat{P}'_1$. The input for e_2 is

$$C\begin{bmatrix}x'^*\\y'^*\end{bmatrix}.$$

C is chosen to give $I_2=I_1=1$. Since we are free to choose its phase, we set it real: $C=1/\sqrt{I'_1}$. The output of e_2 is

$$C_2\begin{bmatrix}x^*\\y^*\end{bmatrix}=\hat{\underline{P}}_1^T\bar{\underline{S}}C\begin{bmatrix}x'^*\\y'^*\end{bmatrix}; \tag{A7}$$

we cannot assume C_2 is real. In fact, using the property (A3) of $\hat{P}_1$, we can write

$$C_2\begin{bmatrix}x^*\\y^*\end{bmatrix}=\begin{bmatrix}x^*\\y^*\end{bmatrix}(x\ \ y)\bar{\underline{S}}C\begin{bmatrix}x'^*\\y'^*\end{bmatrix}$$

(thus fixing C_2) or

$$\begin{aligned}I'_2&=C_2^*C_2\\&=C^2(x'\ \ y')\bar{\underline{S}}^\dagger\begin{bmatrix}x^*\\y^*\end{bmatrix}(x\ \ y)\bar{\underline{S}}\begin{bmatrix}x'^*\\y'^*\end{bmatrix}\\&=\frac{1}{I'_1}(x'\ \ y')\bar{\underline{S}}^\dagger\hat{\underline{P}}_1^T\bar{\underline{S}}\begin{bmatrix}x'^*\\y'^*\end{bmatrix}.\end{aligned} \tag{A8}$$

At this point we are prepared to obtain the two halves of the relation POR* ⟺ ORR.

1. ORR ⟹ POR*

We now assume the ORR. Then

$$\begin{aligned}I'_2&=\frac{1}{I'_1}(x'\ \ y')\underline{S}^*\hat{\underline{P}}_1^T\underline{S}^T\begin{bmatrix}x'^*\\y'^*\end{bmatrix}\\&=\frac{1}{I'_1}\left[(x'\ \ y')\underline{S}^*\begin{bmatrix}x^*\\y^*\end{bmatrix}\right]\left[(x\ \ y)\underline{S}^T\begin{bmatrix}x'^*\\y'^*\end{bmatrix}\right].\end{aligned} \tag{A9}$$

Now we note that I'_1 is a real scalar. The first quantity in square brackets in Eq. (A9) is [cf. Eq. (A5)] $(I'_1)^T$ and the second is I'^*_1, both of which are thus I'_1. So we get

$$I'_2=I'_1 \tag{A10}$$

by assuming the ORR. Thus ORR ⟹ POR*.

2. POR* = > ORR

We now back up to Eq. (A8) and assume instead that the POR* is true. This gives

$$(I'_1)^2=(x'\ \ y')\bar{\underline{S}}^\dagger\hat{\underline{P}}_1^T\bar{\underline{S}}\begin{bmatrix}x'^*\\y'^*\end{bmatrix}. \tag{A11}$$

We also know that

$$\begin{aligned}(I'_1)^2&=(x'\ \ y')\underline{S}^*\begin{bmatrix}x^*\\y^*\end{bmatrix}(x\ \ y)\underline{S}^T\begin{bmatrix}x'^*\\y'^*\end{bmatrix}\\&=(x'\ \ y')\underline{S}^*\hat{\underline{P}}_1^T\underline{S}^T\begin{bmatrix}x'^*\\y'^*\end{bmatrix}.\end{aligned} \tag{A12}$$

The fact that $(x'\ \ y')$ is arbitrary (and complex), plus Eqs. (A11) and (A12), is sufficient to ensure that

$$\bar{\underline{S}}^\dagger\hat{\underline{P}}_1^T\bar{\underline{S}}=\underline{S}^*\hat{\underline{P}}_1^T\underline{S}^T. \tag{A13}$$

This in turn is sufficient to give

$$\bar{\underline{S}}=e^{i\phi}\underline{S}^T, \tag{A14}$$

i.e., we now have the ORR to within an undetermined but global phase. The result (A14) was also obtained by Shelankov and Pikus[15] starting from the weak POR. We repeat their interferometric argument (slightly generalized) to complete the derivation. We surround our system $\underline{S}$ with a set of partial mirrors, which bring about multiple passages through $\underline{S}$ en route from **r** to **r**′ (and vice versa in the modified e_2). The resulting system, which we call $\underline{S}'$ is given by

$$\underline{S}'=s_2(1+r\underline{S}+r^2\underline{S}^2+\cdots)\underline{S}s_1, \tag{A15}$$

where s_1 is the amplitude to enter the first mirror from **r**, $s2$ the amplitude to pass through the second to **r**′, and r the amplitude to be reflected so as to reenter $\underline{S}$ from the direction of **r** (we take all of these to be real). Also

$$\bar{\underline{S}}'=s_1(\underline{1}+r\bar{\underline{S}}+r^2\bar{\underline{S}}^2+\cdots)\bar{\underline{S}}s_2. \tag{A16}$$

If we now set $\bar{\underline{S}}'=e^{i\phi'}(\underline{S}')^T$ and $\bar{\underline{S}}=e^{i\phi}\underline{S}^T$, then Eqs. (A15) and (A16) suffice to give $\phi'=\phi=0$. Thus we have POR* = > ORR.

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$$\varepsilon_{\rm av}=\left[\frac{1}{2}\right]\left[\frac{|T_{++}+T_{+-}|-|T_{-+}+T_{--}|}{|T_{++}+T_{+-}|+|T_{-+}+T_{--}|}+\frac{|T_{++}-T_{+-}|-|T_{-+}-T_{--}|}{|T_{++}-T_{+-}|+|T_{-+}-T_{--}|}\right],$$

while

$$\varepsilon_{\rm CD}=[(|T_{++}|-|T_{--}|)/(|T_{++}|+|T_{--}|)]$$

for the same inputs.
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