Optical non-reciprocity in magnetic structures related to high-$T_c$ superconductors

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Abstract

Recent neutron scattering [1, 2], and optical measurements [3, 4] have detected evidence in underdoped cuprate superconductors for a phase transition near the pseudogap onset temperature $T^*$ to a time reversal-breaking state. The neutron scattering indicates antiferromagnetic ordering, while it is often assumed that optical polarization rotation requires at least a weak ferromagnetic component. In this note we identify several antiferromagnetic structures, compatible with neutron scattering data, that allow intrinsic polarization rotation through the magnetoelectric effect.
High-$T_c$ superconductivity in the cuprates remains a controversial field after more than 20 years of intense research \cite{5}. One of the few ideas that has achieved broad consensus is that solving the puzzle of high-$T_c$ requires first understanding the normal phase from which it evolves. Early in the study of these materials, various probes, particularly thermodynamic ones, failed to detect a phase transition above the critical temperature for superconductivity. Instead, the most prominent feature of the normal state is the loss with decreasing temperature of states at the Fermi energy - the pseudogap phenomenon \cite{6}. The lack of evidence for a phase transition suggested that the opening of the pseudogap reflects a crossover, rather than the appearance of a new phase with a distinct symmetry. Subsequently, careful experiments revealed that in certain cuprates, at doping levels close to $1/8$, there were indeed phase transitions to states with true charge and spin density order \cite{7}. Thus the possibility exists that proximity to these translational symmetry breaking phases controls the physics in cuprates that do not manifest this form of static order.

More recently, striking results have been obtained from neutron scattering experiments \cite{1, 2} on a variety of underdoped cuprates that point to a well-defined phase transition to a state with broken time reversal symmetry. The phase transition occurs at temperatures associated with the appearance of the pseudogap state as determined by probes such as ARPES, NMR, and optical conductivity \cite{6}. Magnetic neutron scattering appears at wavevectors that coincide with reciprocal lattice vectors, indicating that time-reversal breaking occurs without loss of translational symmetry. Furthermore, no scattering is observed when the in-plane component of the scattering vector is zero, suggesting that the net moment of the unit cell is zero. Another significant finding is that the moments are not perpendicular to the Cu-O plane, but are are canted at an angle of about 45 degrees. From these observations it can be concluded that antiferromagnetically-aligned magnetic moments exist within each unit cell, whose origin could be the orbital currents proposed by Varma \cite{8}, electron spins, or some combination of the two.

In another set of elegant experiments \cite{3, 4} performed on cuprate superconductors, the Kapitulnik group has detected a very small difference in the reflection coefficient of left and right circularly polarized light (Kerr effect) using an ultrasensitive Sagnac interferometer. Kerr rotation is a direct manifestation of broken time-reversal, and therefore its appearance tends to support the neutron scattering findings. Although the experiments have yet to be
performed on the same sample, it seems that the onset temperature of the Kerr rotation, \( T_K \), is systematically lower than the onset of antiferromagnetic ordering, \( T_{AF} \), as observed by neutron scattering. One possible explanation is that the Kerr rotation results from "weak ferromagnetism," a net moment induced in a pair of oppositely oriented spins by a small antisymmetric canting of the spin direction. It is possible that weak ferromagnetism sets in at a temperature below \( T_{AF} \), triggered by the onset of some form of charge ordering, such as formation of a nematic state [9–14].

The purpose of this note is to discuss a different, intrinsic, mechanism that generates Kerr rotation without requiring weak ferromagnetism. This mechanism requires that time-reversal symmetry (T) and inversion symmetry (I) are broken, although the product T*I may be a symmetry operation. This is in fact the symmetry of a pattern of intra-unit cell loop currents described by Simon and Varma [15]. On the other hand, if the moments are associated with spins that reside on the O atoms, inversion symmetry is not broken and this mechanism does not operate. As we will see below, the proposed mechanism also fails to operate if the moments are aligned parallel to the normal vector of the Cu-O plane, as predicted in the basic form of the loop-current theory. What is required is that the moments be tipped from the normal, as is in fact observed experimentally.

As pointed out previously [16], a state in which T and I broken are broken by intra-unit cell currents can be in the class of materials that exhibit magnetostrictive phenomena. In these materials generation of magnetization by electric fields, and electrical polarization by magnetic fields, is allowed by symmetry. In a classic experiment, Krichevtskov et al. [17] observed Kerr rotation in the model magnetostrictive antiferromagnet \( \text{Cr}_2\text{O}_3 \). This rotation is referred to as non-reciprocal, meaning that it changes sign in domains in which all the spins are flipped, i.e. the sense of time is reversed. The existence of non-reciprocal optical rotation in magneto-taxtromagnetic materials such as \( \text{Cr}_2\text{O}_3 \) had been predicted theoretically many years ago [18, 19], although the magnitude was thought to be extremely small. Experiments showed that the rotation is reasonably large and readily observable, of order \( 10^{-4} \), illustrating the well-known fact that symmetry arguments are quite powerful in predicting the existence of exotic effects, but not necessarily their magnitude. More recently, microscopic theories that include spin-orbit coupling have brought theory and experiment into agreement [20–22].

Motivated by these experiments, there has been considerable progress in developing a
macroscopic theory of reflection from magnetoelectrics \cite{23,24}. As a result of this work, there exists a theoretical approach to calculate reflection coefficients from macroscopic property tensors that is consistent with the requirements imposed by time-reversal and inversion symmetries \cite{25,26}. Below, we use the theoretical approach described in Ref. \cite{23} to calculate the reflection matrix for several antiferromagnetic structures that appear consistent with the neutron scattering data as it currently exists. Of these structures, it emerges that several give rise to a non-zero Kerr effect. A phase transition from a Kerr-inactive to Kerr-active antiferromagnetic state at $T_K$ is a scenario that could account for the onset of optical rotation at a temperature below the onset of magnetic neutron scattering.

The macroscopic theory of magnetoelectric optics requires consideration of magnetization, $M$, induced by electric fields, $E$, which yields effects at magnetic dipole order. The magnetoelectric tensor $t_{\alpha\beta}$ that relates $M$ to $E$ is a second-rank axial tensor that changes sign under reverse of spin and current, that is a $(c)$ tensor in the notation of Birss \cite{27}. However, it turns out that for the theory to be consistent with time-reversal, electric quadrupole effects must be included as well. A third-rank polar $(c)$ tensor, $S_{\alpha\beta\gamma}$, relates polarization, $P$, to the time and spatial derivative of $E$, yielding effects at electric quadrupole order. Although both $S_{\alpha\beta\gamma}$ and $t_{\alpha\beta}$ contain nonzero elements in all magnetoelectrics, the optical properties of a given material depend sensitively on its magnetic point group.

In the following, we discuss the optical reflectivity associated with five inversion breaking magnetic structures that are relevant to experiments. The first, sketched in Fig. 1(a), is the structure described in Ref. \cite{15}, which belongs to the magnetic point group $mmm'$. This structure is characterized by three mirror planes, two of which contain the Cu-O unit cell diagonals and the normal vector. The third lies in the Cu-O plane. Of the three, the plane defined by the diagonal that passes through the moments and the normal direction is an $m'$ plane, that is, the symmetry operation is mirror followed by reversal of the direction of the orbital currents. As we will see, nonreciprocal rotation is not observable in the $mmm'$ group. This fact, and the evidence from neutron scattering that the moments are not perpendicular to the Cu-O plane, lead us to consider structures with different symmetry. In Fig. 1(b) the magnetic moments are tipped away from the normal direction while remaining within the $m'$ plane. In Fig. 1(c) the moments are again tipped away from the normal, but in the direction perpendicular to the $m'$ plane. The lowering of symmetry removes two of the three mirror
FIG. 1: Representation of possible symmetries of magnetic moments in a planar unit cell. Dashed line represents the Cu-O plane, the plane of drawing contains the normal and one of the diagonals of the CuO₂ cell. (a) moments perpendicular to the plane: point group $mmm$’ (b) moments tipped in the plane of the sketch: point group $2/m'$ (c) moments tipped out of the plane of the sketch: point group $2'/m$.

FIG. 2: Representation of possible symmetries of magnetic moments in a unit cell containing two apical oxygen atoms. (a) point group $mmm$’ (b) point group $m'm'm'$.

planes in both cases. In structure shown in Fig. 1(b) the $m'$ plane remains, as does a 2-fold rotation about the normal to this plane. Thus it is associated with the magnetic point group $2/m'$. In contrast, the structure shown in Fig. 1(c) is characterized by the $2'/m$ group, as the $m$ operation remains, together with a 2-fold rotation followed by inversion about the $m$ plane normal.

In another relevant structure studied by Weber et al. [28], the orbital currents flow through the apical O, rather than the Cu atom at the center of the cell. This structure, represented by four moments in Fig. 2(a), is also characterized by the point group $mmm$’ [16], and is therefore not a candidate structure for nonreciprocal rotation. However, reversing
a pair of moments, as in Fig. 2(b), yields a point group $m'm'm'$ structure, which will turn out to allow non-reciprocal optical rotation. Although the moments in Fig. 2(b) lie in a plane, it may be possible that the moments below the Cu-O plane could be rotated by 90 degrees about $z$-axis with respect to the moments above. Such a structure would belong to the $2m'm'$ group, which has the same optical response as $m'm'm'$.

The breaking of I and T allow for additional terms in the constitutive relations that define the optical response of a magnetolectric medium. In the formulation of Graham and Raab [23], these additional terms are,

\begin{align*}
M_\alpha &= t_{\beta\alpha}E_\beta, \\
P_\alpha &= -\frac{1}{6}iS_{\alpha\beta\gamma}\nabla_\gamma E_\beta,
\end{align*}

where,

\begin{equation}
S_{\alpha\beta\gamma} \equiv a_{\alpha\beta\gamma} + a_{\beta\gamma\alpha} + a_{\gamma\alpha\beta},
\end{equation}

and,

\begin{equation}
t_{\alpha\beta} \equiv G_{\alpha\beta} - \frac{1}{3}\delta_{\alpha\beta}G_{\gamma\gamma} - \frac{1}{6}\omega\epsilon_{\beta\gamma\delta}a_{\gamma\delta}\alpha.
\end{equation}

In the above $G_{\alpha\beta}$ is the second-rank axial (c) tensor and $a_{\alpha\beta\gamma}$ is the third-rank polar (c) tensor. These additional terms, together with Maxwell’s equations, lead to modifications of the boundary conditions and wave impedance. We consider reflection at normal incidence, which we define as the $z$-direction. The boundary condition on the tangential components of $H$ are,

\begin{equation}
\epsilon_{az\gamma}(H_{2\gamma} - H_{1\gamma}) = \frac{1}{5}\omega S_{\alpha\gamma z}E_{1\gamma},
\end{equation}

while the condition of continuity of the tangential component of $E$ remains unchanged. The wave impedance, or coupling between $E$ and $H$ in the medium, is described by the relation,

\begin{equation}
H_\alpha = (\mu_0c)^{-1}n\epsilon_{az\gamma}E_\gamma - t_{\beta\alpha}E_\beta.
\end{equation}

The coupling of $H_x$ to $E_x$ and $H_y$ to $E_y$ via a time-reversal odd tensor is the key ingredient that gives rise to non-reciprocal rotation, as this is similar to the coupling in the presence of an external magnetic field parallel to the $z$ axis. Such coupling arises from Eq. 6 if the
diagonal elements $t_{xx}$ and $t_{yy}$ are nonzero, or from Eq. 5 if the off diagonal components $S_{xyz}$ and $S_{yxz}$ are nonzero. To see which, if any, of the relevant magnetic structures generate a non-reciprocal rotation, we evaluate tensor elements for the magnetic point groups under consideration,

$$m'm': t_{xx} = t_{yy} = t_{xy} = 0; S_{xyz} = S_{yxz} = 0, \quad 7$$

$$2'/m: t_{xx} = t_{yy} = 0; t_{xy} \neq 0; S_{xyz} = S_{yxz} = 0, \quad 8$$

$$m'm'm', 2m'm': t_{xx}, t_{yy} \neq 0; t_{xy} = 0; S_{xyz}, S_{yxz} \neq 0, \quad 9$$

$$2/m': t_{xx}, t_{yy} \neq 0; t_{xy} = 0; S_{xyz}, S_{yxz} \neq 0. \quad 10$$

From the above it is clear that non-reciprocal rotation can be observed in $2/m', m'm'm', 2m'm'$, but not in the other structures.

From the tensors $S_{\alpha\beta\gamma}$ and $t_{\alpha\beta}$, the reflectivity matrix, defined by $E_{r\alpha} = r_{\alpha\beta}E_{i\beta}$ (where subscripts $i$ and $r$ denote incident and reflected respectively), can be derived in the usual way from the boundary conditions and wave impedance relations. The reflectivity matrix for the Kerr-active structures is,

$$r = \begin{bmatrix}
\frac{n-1}{n+1} & \frac{2(-\tau_{yy}+s_{xy})}{(n+1)^2} \\
\frac{2(\tau_{xx}+s_{yx})}{(n+1)^2} & \frac{n-1}{n+1}
\end{bmatrix}, \quad 11$$

where $\tau_{xx} \equiv \mu_0ct_{xx}$, $\tau_{yy} \equiv \mu_0ct_{yy}$, $s_{xy} \equiv (\omega\mu_0c/6)S_{xyz}$, $s_{yx} \equiv (\omega\mu_0c/6)S_{xyz}$. In the above, we have made the approximation that the diagonal elements of $r_{\alpha\beta}$ are equal, as we consider a medium that would be tetragonal if not for the magnetic structure.

The beauty of the Sagnac technique is that it is sensitive only to the time-reversal breaking parameter, $\phi \equiv (r_{xy} - r_{yx})$. For the point groups discussed above, $\phi = -2(\tau_{xx} + \tau_{yy} + s_{xy} - s_{yx})/(n+1)^2$. From the permutation symmetry the third-rank polar tensor $a_{\alpha\beta\gamma} = a_{\alpha\gamma\beta}$ it follows that $s_{xy} - s_{yx} = 0$. Therefore $\phi \propto (\tau_{xx} + \tau_{yy})$, yielding the sensible result that the Sagnac signal is independent of the choice of crystallographic axes. Finally, we note that the reflectivity matrix (Eq. 11) is the same as obtained by Dzyaloshinskii in the context of anyon theory, for the case of a bilayer cuprate in which the gauge fields of the two layers have opposite sign. This state is equivalent to a pair of antiferromagnetically aligned moments displaced along the $z$ axis, instead of the $x$ or $y$ axis as in Fig. 1(a). That the response is
equivalent is consistent with symmetry considerations, as the corresponding magnetic point group is \( m'm'm' \), i.e. the same as in Fig. 2(b).

Although the reflectivity matrix above will yield a Kerr effect, there are several issues that arise when comparing Sagnac measurements with non-reciprocal rotation in magneto-electrics. One is the relatively small size of the effect, of order 0.1-1 microradians in cuprates \[8\] as compared with 100 microradians in \( \text{Cr}_2\text{O}_3 \) \[17\], and a second, perhaps related issue is "training," or alignment of domains. As \( \tau_{xx} + \tau_{yy} \) changes sign under reversal of spin direction, the optical rotation of a multidomain sample averages to zero. The Sagnac signal is found to increase when the sample is cooled in a magnetic field, an effect normally associated with the alignment of ferromagnetic, not antiferromagnetic, domains. However, although antiferromagnetic magneto-electrics do not have free energy terms proportional to a static field in the \( z \)-direction, \( H_z^0 \), there are terms proportional to \( H_z^0 E_z^0 \) in the Kerr-active point groups. If breaking of symmetry at the crystal surfaces generates an \( E_z^0 \), the antiferromagnetic domains near the surface will be aligned by \( H_z^0 \). If the rotation is confined to a thin surface layer the relatively small size of the observed rotation could be explained. Finally, a prediction that distinguishes magneto-electric from ferromagnetic training is that domains on the two opposite sides of the crystal are time-reversed in the magneto-electric case.

To conclude, we have identified several magnetic structures that manifest non-reciprocal optical rotation in the absence of weak ferromagnetism and are relevant to recent neutron scattering results. This finding is independent of whatever interactions generate such structures and whether they arise from spin, orbital angular momentum, or both. In carrying out the symmetry analysis we have considered the highest symmetry (single-layer tetragonal) structure exemplified by the Hg 1201 compound. Extending the analysis to bilayer cuprates is more difficult, as it requires additional assumptions regarding the magnetic coupling between the two layers. However, if Kerr-active antiferromagnetic structures exist in the highest symmetry crystal structure, they will be found in crystals with lower symmetry as well.

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