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

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Abstract

Rotation of the plane of polarization of reflected light (Kerr effect) is a direct manifestation of broken time reversal symmetry and is generally associated with the appearance of a ferromagnetic moment. Here I identify magnetic structures that may arise within the unit cell of cuprate superconductors that generate polarization rotation despite the absence of a net moment. For these magnetic symmetries the Kerr effect is mediated by magnetoelectric coupling, which can arise when antiferromagnetic order breaks inversion symmetry. The structures identifed are candidates for a time-reversal breaking phase in the pseudogap regime of the cuprates.

High-T_c superconductivity in the cuprates remains a controversial field after more than 20 years of intense research [1]. One of the few ideas to achieve 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 [2]. 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 [3]. 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 spin-flip neutron scattering (SFNS) experiments [4, 5] on a variety of underdoped cuprates that suggest a phase transition to a state with broken time reversal symmetry. The onset of SFNS occurs at temperatures associated with the appearance of the pseudogap state as determined by probes such as ARPES, NMR, and optical conductivity [2]. The scattering vectors 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 vanishing of the total moment of the unit cell. Another finding is that the moments are not perpendicular to the Cu-O plane, but are canted at an angle of about 45 degrees. These observations suggest that antiferromagnetically-aligned magnetic moments exist within each unit cell, whose origin could be orbital currents, for example those proposed by Varma [6], electron spins, or some combination of the two.

In view of their potential significance, it is clearly important to seek independent confirmation of the SFNS results. However, at present, the experimental picture is far from clear. Carefully designed NMR [7] and NQR [8] experiments on $YBa_2Cu_4O_8$ place upper bounds on the strength of local magnetic fields that are inconsistent with the moments deduced from SFNS. On the other hand, the Kapitulnik group has used a Sagnac interferometer to detect a difference in the reflection coefficient of left and right circularly polarized light

(Kerr effect) in $YBa_2Cu_3O_{6+x}$, which is direct manifestation of broken time-reversal [9, 10]. Although a non-zero Kerr effect appears to support the SFNS conclusions, important issues remain unresolved. It appears that the onset temperature of the Kerr rotation, T_K, is systematically lower than the ordering temperature, T_{AF}, as observed by SFNS. Furthermore, the Kerr effect is traditionally associated with ferromagnetism, whereas SFNS is detecting antiferromagnetic order. This discrepancy can be explained by "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 [11–16].

The purpose of this Letter is to present a mechanism that generates Kerr rotation in an antiferromagnetically ordered system, 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. Many antiferromagnetic spin systems possess this property [17], as does a class of intra-unit cell loop currents described by Simon and Varma [18]. On the other hand, if the moments are associated with spins that reside on the O atoms, inversion symmetry is not broken and zero Kerr rotation is predicted. As we will see below, the Kerr effect is zero as well if the moments are aligned normal to the Cu-O plane, as predicted for planar loop-currents. What is required is that the moments be tipped from the normal, as the SFNS data indicates.

As pointed out previously [19], a state in which T and I broken are broken by intra-unit cell currents can be in the class of materials that exhibit magnetoelectric phenomena. In these materals generation of magnetization by electric fields, and electrical polarization by magnetic fields, is allowed by symmetry. In a classic experiment, Krichevtsov et al. [20] observed Kerr rotation in the model magnetoelectric antiferromagnet Cr_2O_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 magnetoelectrics such as Cr_2O_3 had been predicted theoretically many years ago [21, 22], 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 [23–25].

Motivated by these experiments, there has been considerable progress in developing a macroscopic theory of reflection from magnetoelectrics [26, 27]. 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 [28, 29]. Below, we use the theoretical approach described in Ref. [26] to calculate the reflection matrix for several antiferromagnetic structures that appear consistent with the NS 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 NS.

The macroscopic theory of magnetoelectric optics requires consideration of magnetization, M, induced by electric fields, E, which yields effects at magnetic dipole order. The magneto-electric 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 [30]. 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. [18], 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 NS that the moments are not perpendicular to the Cu-O plane, lead us to consider structures with different symmetry. In Fig. 1(b) the

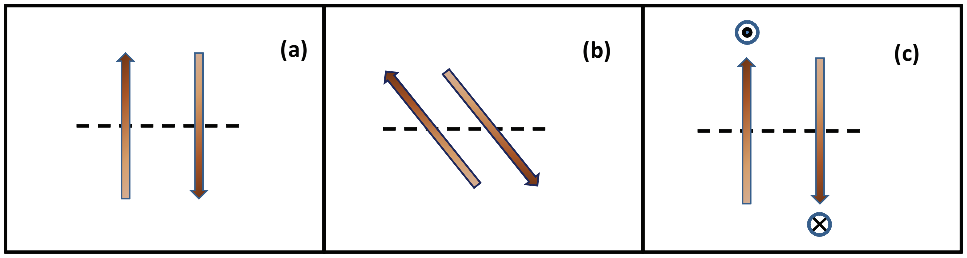

FIG. 1: Representation of possible symmetries of magnetic moments in a planar unit cell. Dashed line represents the Cu-O plane, the plane of drawing contains the normal and one of the diagonals of the CuO_2 cell. (a) moments perpendicular to the plane: point group mmm' (b) moments tipped in the plane of the sketch: point group $2/m'$ (c) moments tipped out of the plane of the sketch: point group $2'/m$.

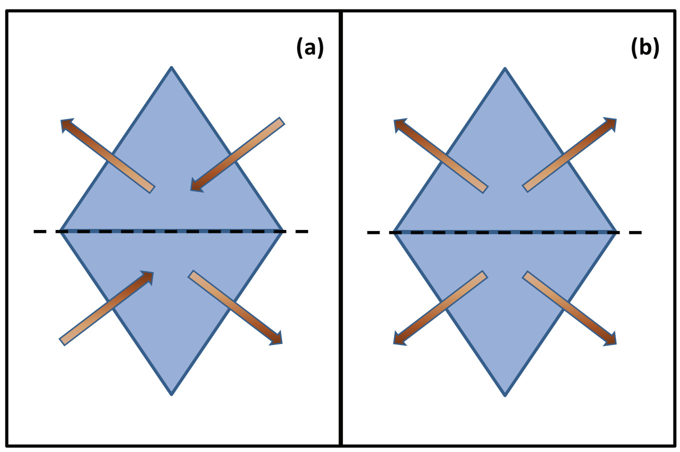

FIG. 2: Representation of possible symmetries of magnetic moments in a unit cell containing two apical oxygen atoms. (a) point group mmm' (b) point group $m'm'm'$.

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 planes in both cases. In the 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.* [31], 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' [19], 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 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 the define of the optical response of a magnetolectric medium. In the formulation of Graham and Raab [26], these additional terms are,

$$M_\alpha = t_{\beta\alpha} E_\beta, \tag{1}$$

$$P_\alpha = -\frac{1}{6} i S_{\alpha\beta\gamma} \nabla_\gamma E_\beta, \tag{2}$$

where,

$$S_{\alpha\beta\gamma} \equiv a_{\alpha\beta\gamma} + a_{\beta\gamma\alpha} + a_{\gamma\alpha\beta}, \tag{3}$$

and,

$$t_{\alpha\beta} \equiv G_{\alpha\beta} - \frac{1}{3}\delta_{\alpha\beta} G_{\gamma\gamma} - \frac{1}{6}\omega\epsilon_{\beta\gamma\delta} a_{\gamma\delta\alpha}. \tag{4}$$

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,

$$\epsilon_{\alpha z\gamma}(H_{2\gamma} - H_{1\gamma}) = \frac{1}{6}\omega S_{\alpha\gamma z} E_{1\gamma}, \tag{5}$$

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,

$$H_\alpha = (\mu_0 c)^{-1} n \epsilon_{\alpha z\gamma} E_\gamma - t_{\beta\alpha} E_\beta. \tag{6}$$

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,

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

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

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

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

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,

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

where $\tau_{xx} \equiv \mu_0 c t_{xx}$, $\tau_{yy} \equiv \mu_0 c t_{yy}$, $s_{xy} \equiv (\omega\mu_0 c/6)S_{xyz}$, $s_{xy} \equiv (\omega\mu_0 c/6)S_{xyz}$. 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 of the third-rank polar tensor $a_{\alpha\beta\gamma} = a_{\alpha\gamma\beta}$ [26] 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 [32] 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 [9] as compared with 100 microradians in Cr_2O_3 [20], 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 magnetoelectrics 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 magnetoelectric from ferromagnetic training is that domains on the two opposite sides of the crystal are time-reversed in the magnetoelectric case.

To conclude, we have identified several antiferromagnetic structures that manifest non-reciprocal optical rotation and are candidates for a time-reversal breaking phase in cuprate superconductors. 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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