Gyrotropic birefringence in the underdoped cuprates

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Abstract – The optical effects due to the loop-current order parameter in underdoped cuprates are studied in order to understand the recent observation of unusual birefringence in electromagnetic propagation in underdoped cuprates. It is shown why birefringence occurs even in samples with multiple domains of size much smaller than the wavelength and in twinned samples. Not only is there a rotation of polarization of incident light but also a rotation of the principal optical axis from the crystalline axes. Both are calculated in relative agreement with experiments in terms of the same parameters. The magnitude of the effect is orders of magnitude larger than the unusual Kerr effect observed in underdoped cuprates earlier. The new observations, including their comparison with the Kerr effect, test the symmetry of the proposed order decisively and confirm the conclusions from polarized neutron scattering.

Introduction. – The heart of the solution of the cuprate problem is in ascertaining the physics of the strange-metal phase [1] and the so-called pseudo-gap phase [2,3]. One theoretical approach [4–6] suggests that the strange-metal region of the cuprates, as well their high temperature $d$-wave superconductivity, is due to the scattering of fermions from the quantum-critical fluctuations of an unusual time-reversal breaking phase, which has a finite magneto-electric tensor and which occupies the so-called pseudo-gap region of the phase diagram. Various experiments [7–9] have provided evidence for a broken symmetry at the onset of the pseudo-gap region. Polarized neutron scattering in four families of cuprates [10] and dichroic ARPES [11] in one have provided direct evidence of the predicted symmetry. Controversy, some of it based on scientific grounds [12–14], however continues. It is therefore of great interest that experiments [15] with quite a different technique are now available to address the issue of the phase transition to the pseudo-gap phase and the specific symmetry of the magneto-electric tensor proposed for it.

These experiments are optical experiments [15] which have observed birefringence with several unusual features in underdoped cuprates below the temperatures $T^*(x)$ at which other signatures of the pseudo-gap are observed. The fact that they begin to be observed below $T^*(x)$ and their magnitude grows below it indicates of course that $T^*(x)$ marks a new symmetry. But the experiments are done in twinned films so that birefringence would normally not be observed. Even in single crystals, no birefringence can be usually observed if there is a symmetry breaking but with domains of different equivalent order of size much smaller than the optical wavelength. The other remarkable features of the observations are that the principal axes of the birefringence are not the crystalline axes but those rotated by an angle $\theta_P$ with respect to them. Another feature is that the angle of rotation of the polarization $\theta_R$ and $\theta_P$ defined with respect to the propagation direction remains the same on shining light on the two opposite faces of the sample with respect to the $ab$ planes. In materials with time-reversal breaking, some unusual optical effects have been predicted [16] and some observed but none of the kind discovered in cuprates. I show here that the observations follow from the symmetry of the magneto-electric tensor proposed to occur in the cuprates. I also suggest further experiments to verify some untested aspects of the results obtained here. I will also compare the difference in the occurrence and magnitude of the unusual Kerr effect [17,18] observed earlier in underdoped cuprates compared to the recent birefringence measurements. There is much to be learnt from this comparison.

The general optical effects in materials with magneto-electric symmetry and with application to the symmetry of $\text{Cr}_2\text{O}_3$ and $\text{MnTiO}_3$ were proposed by Brown et al. [16]; the corresponding Maxwell equations were written down by Hornreich and Shtrikman [19]. I write them here for the propagation of light in a symmetry appropriate to that proposed for the cuprates and provide a physical explanation for the unusual features.
\textbf{Analysis.} – Let us start with the Maxwell equations:

\begin{equation}
ce_{ijk}E_qq_k = -\omega B_i, \quad ce_{ijk}H_jq_k = \omega D_i,
\end{equation}

where \( e_{ijk} \) is the totally antisymmetric unit matrix. The material properties are defined through the relations

\begin{align}
D_i &= \epsilon_{ij}E_j + \chi_{ij}^{EM}H_j, \\
B_i &= \chi_{ij}^{ME}E_j + \mu_{ij}H_j,
\end{align}

\( \chi_{ij}^{EM} \) and \( \chi_{ij}^{ME} \) are the elements of the magneto-electric tensor. It is assumed that there is no natural optical activity in the material, \( i.e. \) the material is not dielectrically chiral.

Following Agranovich and Ginzburg [20], one can combine all induced effects in a generalized polarizability \( P'_i \) rather than introducing them separately in an induced magnetization by defining

\begin{equation}
P'_i = P_i + (c/\omega)e_{ijk}M_jq_k, \quad D'_i = E_i + 4\pi P'_i
\end{equation}

so that the Maxwell equations can be written in terms of \( D'_i \) and \( B_i = H_i \). The effective polarization \( D'_i \) may then be written with \( (i, j, k) \) taken as the principal axes such that the ordinary dielectric matrix \( \epsilon_{ij} \) and the permittivity matrix \( \mu_{ij} \) are diagonal, with values \( \epsilon_i \) and \( \mu_i \), respectively. Then

\begin{equation}
D'_i = (\epsilon_{ij} + \gamma_{ijk}q_k - (c/\omega)^2(1 - \mu_i^{-1})\epsilon_i\delta_{ij}(1 - \delta_{ik})) E_j,
\end{equation}

where the renormalized susceptibility and gyrotropic tensors are

\begin{align}
\bar{\epsilon}_{ij} &= \epsilon_{ij} - \chi_{ik}^{EM}\mu_k^{\bar{\epsilon}}\chi_{kj}^{ME}, \\
\bar{\gamma}_{ijk} &= i(c/\omega)(\epsilon_{ij}\chi_{ik}^{EM} + \epsilon_{jk}\chi_{il}^{EM})\mu_i^{-1}.
\end{align}

\textbf{Effects in the specific symmetry of the loop-ordered state.} – We now consider the symmetry of the proposed state in underdoped cuprates where an order parameter exists so that an effective free-energy term is \( \bar{\epsilon}_{ij}^{\ast} \chi_{ij}^{ME} \varphi_{i}^{\ast} \varphi_{j} \), where \( \chi_{ij}^{ME} \) are finite. This order parameter arises due to orbital currents which have current loops in each unit cell as depicted in fig. 1. The order parameter is characterized by the anapole vector

\begin{equation}
\Omega = \int_{\text{cell}}d^2r(M(r) \times r).
\end{equation}

In a tetragonal crystal the symmetry class is \((mm\overline{2}m)\) [21]. I will assume a tetragonal symmetry even in orthorhombic crystals for calculation of the extra effects due to the loop current order because the corrections due to the orthorhombic symmetry are small. (We must however keep track of the ordinary birefringence of the orthorhombic symmetry.) There are four domains, two with anapole vectors oriented in the \( \pm \hat{x}' = \pm (1/\sqrt{2})(\hat{x} + \hat{y}) \) directions, and two with anapole vectors oriented in the \( \pm \hat{y}' = \pm (1/\sqrt{2})(\hat{x} - \hat{y}) \) directions. The former have only non-zero real elements \( \chi_{xx,xx}^{EM} = \chi_{xx,xx}^{ME} \), with equal values for the two domains. Similarly the latter have only non-zero real elements \( \chi_{yy,yy}^{EM} = \chi_{yy,yy}^{ME} \). Also \( \chi_{xy,xy}^{EM} = \chi_{xy,xy}^{ME} \). These are evident already from the cartoons in fig. 1.

We will consider here only the propagation in the \( z \)-direction: \( k = k_z \). In this case the relevant non-zero off-diagonal elements of the renormalized permittivity are

\begin{equation}
\bar{\epsilon}_{xy} = \mu_x^{-1} \chi_{xx,xy}^{ME} = \bar{\epsilon}_{yx}.
\end{equation}

There is also an equal correction to the diagonal parts \( \bar{\epsilon}_{xx} \) and \( \bar{\epsilon}_{yy} \) which may be ignored due to the much larger ordinary parts. The only components of \( \gamma \) that are relevant for the \( \pm \) axis propagation are \( \gamma_{xz} \) and \( \gamma_{yz} \). From eq. (6), it follows that they are 0.

The occurrence of an off-diagonal component in the dielectric permittivity \( \bar{\epsilon}_{xy} \), from which all effects derived here follow, starting from a magneto-electric tensor whose components are \( \chi_{xx,xy}^{ME} \), may be understood as follows. The free-energy contains terms of the form \( \chi_{xx,xy}^{ME}H_zE_x + \chi_{xy,xy}^{ME}H_yE_y \) (plus similar terms with \( x \rightarrow y \)) as well as a term \( 1/2\mu_x^{-1}H_z^2 \). On propagation of light along the \( z \)-axis with an electric field in the \( x \)-direction generates a magnetic field in the \( z \)-direction which may be eliminated such that an effective-free energy term is \( \bar{\epsilon}_{xy}E_xE_y \) obtained with \( \bar{\epsilon}_{xy} = \chi_{xx,xy}^{ME}H_z^{-1} \chi_{xy,xy}^{ME} \). This quadratic dependence of \( \bar{\epsilon}_{xy} \) on the order parameter plays an important role in what follows.

Inserting eq. (6) in (5) and considering the transverse propagation alone and using the Maxwell equations (1),

\begin{align}
D'_x &= (c/\omega)^2k_x^2E_x = \\
&= (\epsilon_{xx} + (c/\omega)^2k_x^2(1 - \mu_x^{-1}))E_x + (\bar{\epsilon}_{xx})E_y, \\
D'_y &= (c/\omega)^2k_y^2E_y = \\
&= (\epsilon_{yy} + (c/\omega)^2k_y^2(1 - \mu_y^{-1}))E_y + (\bar{\epsilon}_{yy})E_x.
\end{align}

The usual refractive indices are given by \( n_{ax}^2 = \epsilon_x\mu_x, \quad n_{by}^2 = \epsilon_y\mu_y \). Rewrite eqs. (10), (11) in terms of the allowed refractive indices \( n \equiv (c/k_x) \) and \( n_{ax}^2 \equiv \mu_x\bar{\epsilon}_{xx}, \quad n_{by}^2 \equiv \mu_y\bar{\epsilon}_{xy} \);

\begin{align}
n_{ax}^2E_x + n_{by}^2E_y &= 0, \\
n_{by}^2E_x + n_{ax}^2E_y &= 0.
\end{align}
Equations (12), (13) give that the solution for the $n$'s is
\[
(n_1^2, n_2^2) = 1/2(n_{0x}^2 + n_{0y}^2) \\
\pm 1/2\sqrt{(n_{0x}^2 - n_{0y}^2)^2 + 4\Lambda^4},
\]
where $\Lambda^2 = n_{xy}n_{yx}$. The eigenvectors of (12), (13) have the property that
\[
(E_x/E_y)_1 = \Lambda^2/(n_1^2 - n_{0x}^2), \quad (E_y/E_x)_1 = \Lambda^2/(n_2^2 - n_{0y}^2).
\]
These specify the two principal optical axes, i.e. when the incident polarizations have these ratios, there is no dichroism. For all other angles there is.

Now we may explore the consequences of eqs. (14), (15). The first important result is that since $\Lambda^4$ is proportional to $\varepsilon_{xy}^2$, the results are independent of the domains of the order in general. Some specific results are different for single crystals and poly-crystalline materials, as described below. The former must further be divided into a single-domain order (which may be hard to obtain) or an order with many domains in the field of vision.

**Single crystals.** – It follows from (15) that in a tetragonal crystal, i.e. $n_{0x} = n_{0y} = n_0$, the principal axes are at $\pi/4, 3\pi/4$ to the crystalline axes. However, the velocity of propagation when the polarization is at any other angle is increased/decreased for the projection to these principal axes,
\[
(c/n_1, c/n_2) \approx \pm c/n_0 \left(1 \mp \Lambda^2/n_0^2\right).
\]
Changing the propagation from $+\hat{z}$-direction to $-\hat{z}$-direction is equivalent to redefining the axes: $\hat{x} \rightarrow \hat{x}; \hat{y} \rightarrow -\hat{y}$, or the other way around. This is equivalent to changing $n_1 \rightarrow n_2$. So, as in ordinary birefringence, their is no rotation in propagation in a given direction and then back along the same path. For an order with multiple domains in single crystals, which is to be expected, the effects remain the same because $|\Lambda^2|$ is the same in all four domains. The switching of the principal axes from the crystalline axes for $T \gtrsim T^*(x)$ to half-way between them for $T \lesssim T^*(x)$ is a strong prediction of the considerations here. As usual effects due to impurities, fluctuations etc., will in general round out the transition.

For an orthorhombic single crystal, eq. (15) gives that the rotation angle of the principal axes $\theta_p \approx \arctan(|\Lambda^2|/(n_{0x}^2 - n_{0y}^2))$. The value of the rotation angle, $\theta_R$ for arbitrary polarization may be directly determined by projecting the initial polarization to the principal axes and noting the change in polarization in the propagation due to the difference in velocities of the two components, (16). So there is both a rotation of the principal axes, $\theta_p \neq 0$ and a rotation of the polarization $\theta_p \neq 0$. These effects remain the same due to the multiple domains of the order in a single orthorhombic crystal because $|\Lambda^2|$ remains the same. Both effects are in general temperature dependent because $\Lambda^2$ increases for $T \lesssim T^*(x)$.

**Twinned samples.** – The available experiments are performed in twinned samples of the orthorhombic compound YBa$_2$Cu$_3$O$_{6+x}$ with twin size much smaller than the wavelength of light. In this case, a suitable averaging procedure must be found. Consider eq. (14). Obviously, in the twinned samples, the first term $1/2(n_{0x}^2 + n_{0y}^2)$ may be averaged to its mean value $\bar{n}^2$. But the second term does not average to zero for $\Lambda^2 \neq 0$. As we will discuss below, the measured $(\delta n)^2 \equiv |n_{0x}^2 - n_{0y}^2|$ is expected to be much larger than $\Lambda^2$. So, we may approximate
\[
(n_1^2, n_2^2) \approx \bar{n}^2 \pm 2\Lambda^2/\delta n^2, \\
E_x/E_y = E_y/E_x = \\
\pm \frac{2|\Lambda^2|}{\sqrt{((\delta n^2)^2 + 4\Lambda^4)}} \approx \pm \frac{2\Lambda^2}{(\delta n^2)^2}.
\]
This gives that the principal axes are at
\[
\mathbf{d}_1 \approx \pm \left(\hat{x} + \frac{2|\Lambda^2|}{(\delta n^2)^2}\hat{y}\right),
\]
to $O(\Lambda^4/(\delta n^2)^2)$, the vector $\mathbf{d}_2$ is orthogonal to it.

Consider the magnitude of the dichroism. It is given in terms of the ratio of the difference of the velocities to the average velocity
\[
\delta v/v \equiv c(1/n_1 - 1/n_2)/(\bar{n}/\bar{n}) \approx \\
1/2 \left(\sqrt{(\delta(n^2)^2 + 4\Lambda^4)}\right)/\bar{n}^2 \approx \frac{\Lambda^4}{(\delta n^2)^2} \frac{1}{\bar{n}^2}.
\]
The angle of rotation (which varies in a fourfold way with respect to the angles $\theta_p$) has as its maximum in units of the angular rotation per travel over a wavelength,
\[
\theta_{Ron} \approx 4\pi \frac{\Lambda^4}{(\delta n^2)^2} \frac{1}{\bar{n}^2} \text{ rad.}
\]
As temperature decreases, the magnitude of birefringence should increase as the fourth power of the loop-current order parameter. In experiments, the dichroism is expressed in terms of a complex angle $\theta_R = \theta_R' + i\theta_R''$. The rotation angle is complex because typically in a metal the real and imaginary parts of the index of refraction are equal because $\epsilon'' \gg \epsilon'$. In experiments $\theta_R' \approx \theta_R'$. This is of course obtained from eq. (20) for such a normal complex refractive index $\bar{n}$.

**Comparison with experiments.** – Let us first consider the unusual qualitative features of the experimental results [15]. 1) A birefringence is observed in a polycrystal below $T \approx T^*(x)$ and it increases in magnitude as temperature decreases, 2) the principal axes are rotated with respect to the crystalline axes, and 3) the effect has the same sign for shining light normally on the opposite basal plane faces of the sample. All these three remarkable effects follow from eqs. (19), (20). They arise because $|\Lambda^2|$ is independent of the switching of the $x$- and the $y$-axis and of the orientation of the domains of $\Omega$ as shown above. Note
that the observed effects test the specific symmetry predicted of the magneto-electric phase. The results are quite different from what is predicted and observed for example in the classic antiferromagnetic-magneto-electric insulator Cr₂O₃ primarily because its magneto-electric tensor is diagonal while it is off-diagonal, as specified above, for the underdoped cuprates. Polarization with an external electric and/or magnetic field to align the domains of the order in a single crystal of Cr₂O₃ are therefore required to observe θₚ and θₐ.

From eq. (20), it follows that, since Λ² is proportional to the square of the order parameter, the rotation angle θₐ should increase as the fourth power of the order parameter. I note that in the experiment, the increase of θ below an approximately determined χ is quadratic but may be fitted as a linear increase but may be fitted as (T*(x) − T)², consistent with the above result for a mean-field value of the order parameter exponent.

The experiments [15] give both the real and imaginary parts of θₐ as a function of temperature and report that at low temperatures θₚ ≈ 10⁸. They do not report the temperature dependence of θₚ. A temperature dependence is predicted by eq. (19). A direct quantitative comparison with the experimental results is hampered by the lack of information on (δn²) at the measurement frequency and by the lack of an absolute determination of Λ². The dimensionless order parameter, the ratio of the imaginary component of the transfer integral induced by time-reversal breaking to the normal real part around a loop, is estimated [5,22] to be O(10⁻¹). But it is not possible to reliably calculate the magnitude of the εᵥ from this (nor am I aware of instances where this kind of thing has been calculated reliably in even much simpler situations where usually only the symmetry of the optical anisotropies are tested in experiments).

We may however relate the parameters required to get the measured value of θₚ at low temperatures to the measured value of θᵞ. We can use eq. (19) to find Λ²/(δn²) ≈ 1/12 for the measured θₚ of about 10⁸ at the lowest temperature. The difference in conductivity [23,24] of the single crystal YBa₂Cu₃O₆.₅ at frequencies down to about 10¹¹ Hz, (an order of magnitude larger than the birefringence measurements), along the crystalline axis, may be read off to conclude that, due to the conductivity of the chains, (δn²)/n² is of O(1). If we use this estimate, we get from (20) that the rotation angle should be about 80 mrad, while the experiments [15] give a maximum of about 60 mrad.

On entering the superconducting state, the magneto-electric tensors retain their symmetry, although their magnitudes would change in principle. Birefringence would remain qualitatively the same but smoothly changes in magnitude, as given by the changed refractive indices and the magnitude of χ’s.

An interesting question relates to which of the two solutions in eq. (17) is adopted for a randomly arranged order for a given angle of incidence with respect to the lab frame. These two solutions give birefringence zeroes at π/2 relative to each other. For less than perfect disorder, the choice in such cases is determined by non-equilibrium considerations of propagation to realize the maximum entropy production, i.e. maximum loss. So angles along which the conductivity is actually poorer will be chosen.

It is worth remarking on what is expected if light is incident on experimental geometries such that it can be incident at angles to the planes. The most interesting limiting case is incidence along the 110 axis, electric polarization along the 110 axis and magnetic field perpendicular to the plane. In this case the electric field and the magnetic field are such that a linear effect due to the symmetries of χEM, χME is expected, similarly to the suggestion in Cr₂O₃ [19]. However, this linear effect cancels due to domains, just as in that case, and there are no effects quadratic in the order parameter. Unlike in the insulating magneto-electric single-crystal Cr₂O₃, the polarization of domains by external fields is not possible in metallic cuprates. Twinning of the sample also makes the effect zero. The same is true for the incidence along the 100 axis which may be resolved into perpendicular components along 110 and 110 axes. For this reason, the geometry that was adopted in the experiment [15] appears to be the best. An additional prediction of the results here is the systematic loss of signal of birefringence for angles of incidence away from the normal.

Finally, I contrast the measurements of the unusual (Kapitulnik) Kerr effect [17,18] in underdoped YBa₂Cu₃O₆+x with the gyrotropic birefringence. Several explanations of the Kerr effect have been proposed [25]. In the single crystals of this compound, Kerr effect occurs starting at a temperature Tₜ₁₁₁₁₁₁(x) < T*(x) (but extrapolating to T → 0 to the quantum-critical point at x_c). The birefringence appears to occur starting at T*(x) measured by transport and by polarized neutrons. It has been shown earlier [26] that the magneto-electric state does not have a Kerr effect, but that given its presence, lattice distortions of certain symmetry necessarily induce an additional loop order which has an anomalous Hall effect and therefore a Kerr effect. Such lattice distortions do appear to occur in YBa₂Cu₃O₆+x at temperatures consistent with Tₜ₁₁₁₁₁₁(x). The magnitude of the Kerr effect then depends on an additional small parameter—the square of the relevant lattice distortion and is therefore expected to be much smaller than the birefringence. Because the experiments are done at quite different frequencies, a direct comparison is not possible. But it should be noted that the angle in similar units is about four orders of magnitude smaller in the Kerr effect. It should also be noted that one of the peculiar features of the Kerr effect is that it is observed at all in a sample expected to have many domains of oriented order, each with size much smaller than the wavelength. Moreover, the direction of rotation is independent of the sample across Tₜ₁₁₁₁₁₁(x) and then measuring on cooling below. These unusual features have been
explained earlier [26] from some of the same features of the loop current order as used above for the birefringence.

The results obtained here can be further tested by doing birefringence experiments in single crystals, both in tetragonal and orthorhombic symmetries and at the frequencies where Kerr effect results are available. In particular a different magnitude of the effects and their temperature dependence is predicted for twinned crystals.

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