Violation of Onsager Reciprocity in Underdoped Cuprates?

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One of the canons of condensed matter physics is the Onsager Reciprocity principle in systems in which the Hamiltonian commutes with the time-reversal operator. Recent results of measurements of the Nernst coefficient in underdoped \( YBa_2Cu_3O_6+x \), together with the measurements of the anisotropy of conductivity and the inferred anisotropy of the thermopower, imply that this principle is violated. The probable violation and its temperature dependence are shown to be consistent with the Loop-current phase which has been directly observed in other experiments. The violation is related directly to the magneto-electric symmetry of such a phase in which an applied electric field generates an effective magnetic field at right angle to it and to the order parameter vector, and vice-versa.

Extraordinary Experimental results for magneto-transport experiments have been recently presented for various dopings in YBCO. Starting below the pseudogap temperature \( T^*(x) \), the magnitude of the Nernst coefficient rises. Persuasive evidence has been presented that except for a narrow region \( \delta T_c/T_c \lesssim 1/4 \), it is due to quasi-particles. We are not concerned here with its overall magnitude, but only with its symmetry. Nernst effect is the chiral response of fermions to mutually orthogonal magnetic field and a thermal gradient. We focus attention on the observation that the observed chirality in Nernst effect is non-reciprocal (NRN); For a magnetic field in the \( \hat{z} \)-direction, the absolute magnitude of its value \( |\nu_{ba}| \) is found to be different with the thermal gradient applied in the \( \hat{b} \) direction and voltage measured in the \( \hat{a} \) direction, from \( |\nu_{ab}| \), the value with the thermal gradient applied in the \( \hat{a} \) direction and voltage measured in the \( \hat{b} \) direction. \( \hat{a} \) and \( \hat{b} \) are the directions of the orthorhombic crystalline axes in the Cu-O planes, \( \hat{b} \) is taken to be along the chains. A suitable definition of the NRN in this case, which avoids issues of sign of the Hall effect, thermopower etc., which are not relevant to the present discussion, is

\[
NRN \equiv \frac{|\nu_{ba}| - |\nu_{ab}|}{|\nu_{ba}| + |\nu_{ab}|} \quad (1)
\]

In Ref. [2], it is concluded that NRN varies smoothly above \( T^*(x) \) but that it has a contribution below \( T^*(x) \) consistent with \( \alpha \propto (T^*(x) - T) \) for small \( (T^*(x) - T)/T^*(x) \), saturating to a constant value at lower temperatures, i.e. it has a contribution similar to the square of an order parameter in the mean-field regime of a phase with transition temperature \( T^*(x) \). This behavior is observed in the range \( 0.1 \lesssim x \lesssim 0.18 \). In these experiments, as well as in others, the uncertainty in determination of \( T^*(x) \) is \( \approx \pm 20K \). We may isolate the temperature dependent contribution below \( T^*(x) \) by defining

\[
X(T) = NRN(T) - NRN(T \geq T^*(x)), \quad T < T^*(x) \quad (2)
\]

Related quantities are deduced in Ref. [2]. The Nernst coefficient, linear in \( B \), under the condition of zero-transverse electrical current is given by \( [3] \)

\[
\nu_{ba} = -\frac{\sigma_{aa}\alpha_{ba} - \alpha_{ba}\sigma_{aa}}{\sigma_{aa}\sigma_{bb}} \quad (3)
\]

In Eq. (3) \( \sigma \) and \( \alpha \) are the conductivity and thermoelectric tensors. For \( \nu_{ab} \), the subscripts, \((a, b) \to (b, a)\).

There are two classes of possibilities for NRN:

(I) Diagonal anisotropy only : \( \sigma_{aa} \neq \sigma_{bb} \) or \( \alpha_{aa} \neq \alpha_{bb} \) or both.

(II) Off-diagonal anisotropies \( \alpha_{ab} \neq -\alpha_{ba} \) or \( \sigma_{ab} \neq -\sigma_{ba} \) or both. This may or may not be accompanied by diagonal anisotropies.

\( YBCO \) is orthorhombic, so anisotropies in the diagonal components of \( \sigma \) and \( \alpha \) are to be expected at all temperatures. The anisotropy has been explicitly measured for the conductivity [2][4], it is weakly temperature dependent and varies quite smoothly across \( T^*(x) \). Explicit results for the anisotropy of thermopower are not available for many compositions. The ratio measured for the \( O_{12} \) compound is slowly varying at all temperatures [3]. Thermopower measured [4] along the \( a \)-direction at various dopings of interest is just as smooth across \( T^*(x) \) as \( \sigma_{aa} \) and quite unlike the Nernst coefficient. In general, it is improbable that thermopower show non-analytic behavior while the conductivity does not. This view is strengthened by the available data [3][5]. Therefore, possibility I can be excluded for \( X(T) \).

Consider possibility II. If the states of the system are eigenstates of the time-reversal operator Onsager Reciprocity relations [1][6] hold so that \( \sigma_{ab} = -\sigma_{ba} \) for any crystalline symmetry. Also, if as is usual the energy relaxation rate is related to the momentum relaxation rate so that the Mott relation is obeyed, \( \alpha_{ab} = -\alpha_{ba} \). So for a state with time-reversal preserved \( NR \) must be smooth across \( T \approx T^*(x) \). The experimental observations therefore imply possible time-reversal violation in the pseudogap phase. However as seen below, this is only necessary, not sufficient; not all time-reversal violating states and crystalline symmetries lead to the observed behavior of \( X(T) \). For example, ferromagnetism or states with sim-
Simple spin or orbital antiferromagnetic symmetries cannot give the observed behavior. The experiments may then be a macroscopic manifestation of time-reversal breaking with rather special features.

Transition to states with time-reversal violation (TRV) were predicted with an onset temperature $T^*(x)$ [7] with symmetries consistent with observations in four different families of cuprates, by polarized neutron scattering [8] or dichroic ARPES [9]. Polar Kerr effect consistent with ferromagnetism with a small moment has also been observed [10] and is discussed in Ref. [11]. We show that such a state in an orthorhombic crystalline symmetry does indeed give a NR in the Nernst effect consistent with the experiments.

The symmetry of the observed TRV state has been discussed in Ref. [7] [12]. The order parameter violates time-reversal and reflection symmetries but preserves translational symmetry and is given by the polar time-reversal odd vector, an anapole-vector,

$$\mathbf{L} = \int_{\text{unit-cell}} d^2r \ (\mathbf{m}(\mathbf{r}) \times \hat{\mathbf{r}}).$$

$\mathbf{m}$'s are the generated orbital magnetizations due to electron-electron interactions below $T^*(x)$. They point along $\pm \hat{\mathbf{z}}$ in two of the opposite O-Cu-O triangles in the unit-cell. One of the four possible domains in a orthorhombic crystal with $\mathbf{m}$ and $\mathbf{L}$ displayed is shown in Fig. [1]. In an orthorhombic crystal, the four domains have

$$\mathbf{L} = \pm L_x \hat{\mathbf{x}} \pm L_y \hat{\mathbf{y}}.$$  

We will take $\hat{\mathbf{x}}, \hat{\mathbf{y}}$ to be along the $a$ and $b$-directions of the crystal respectively. In a tetragonal crystal $L_x = L_y$, but not in an orthorhombic crystal.

As has been noted [7] [12] this state has Magneto-electric symmetry [13]: An external field $B\hat{z}$ generates an electric field, or more generally any vector $\mathbf{r}$ of the symmetry of an electric field (polar- time-reversal even) in the plane; an external electric field $\mathbf{E}$ in the plane generates a magnetic field, or more generally any vector $(r \times \mathbf{v})$ of the same symmetry as a magnetic field (axial-time-reversal odd) along $\hat{\mathbf{z}}$. Such generated vectors in a metal can be represented by fermion operators, so that for $\mathbf{L} \neq 0$, the Hamiltonian acquires terms,

$$H' = \chi \left( (\mathbf{L} \times \mathbf{B}) \cdot \psi^\dagger(\mathbf{r}) \mathbf{e} \cdot \mathbf{r} \cdot \mathbf{r}(\mathbf{r}) \right)$$

$$+ \left( (\mathbf{L} \times \mathbf{E}) \cdot \psi^\dagger(\mathbf{r}) \left( \frac{e(\mathbf{r} \times \mathbf{v} - \mathbf{v} \times \mathbf{r})}{2e^*} \right) \psi(\mathbf{r}) \right).$$

$\psi, \psi^\dagger$ are the fermion annihilation and creation operators and $\chi$ is the magneto-electric coefficient. We have chosen units such that electric and magnetic fields have the same dimensions so that $\chi L$ is dimensionless. We have also introduced a phenomenological velocity $c^*$, which ordinarily would be the velocity of light $c$. However, since the effective electric and magnetic fields are generated through electron-electron interactions, it is quite likely that $c^*$ is related to the microscopic parameters of the model and so much less than $c$. Only a microscopic calculation of the magneto-electric coefficient can tell; at the moment we leave it as a parameter to be determined.

The force on the fermions due to $H'$ is obtained by calculating the commutator with the momentum operator $\mathbf{p}$ to get

$$\frac{d\mathbf{p}}{dt} = e\chi \left( (\mathbf{L} \times \mathbf{B}) - \frac{(\mathbf{L} \times \mathbf{E}) \times \mathbf{v}}{c^*} \right)$$

We see that in addition to the external electric $\mathbf{E}$ and magnetic $\mathbf{B}$ fields, internal electric $\mathbf{e}$ and magnetic $\mathbf{b}$ fields act on the fermions:

$$\mathbf{e} \equiv \chi(\mathbf{L} \times \mathbf{B}), \quad \mathbf{b} \equiv -\chi(c/c^*)(\mathbf{L} \times \mathbf{E}).$$

Standard transport theory [14] now gives that charge-Hall current $j^H_c$ is given by

$$j^H_c = \sigma^H(\mathbf{B} + \mathbf{b}) \times (\mathbf{E} + \mathbf{e}),$$

where $\sigma^H$ is the normal Hall conductivity tensor. Formally, reciprocity is obeyed only for interchange of direction of charge-Hall current and of $(\mathbf{E} + \mathbf{e})$. Looking at [8], this cannot in general be achieved in experiments. Similarly the thermal Hall current is given by

$$j^H_t = \alpha^H(\mathbf{B} + \mathbf{b}) \times (\mathbf{E} + \mathbf{e}),$$

where $\alpha^H$ is the normal thermal Hall conductivity tensor which in the present case formally obeys reciprocity only for interchange of direction of thermal-Hall current and of $(\mathbf{E} + \mathbf{e})$.

FIG. 1: One of the domains of the observed order. A rectangular unit-cell is shown with orbital-magnetic moments in the + and - $\hat{\mathbf{z}}$ directions in two of the four O-Cu-O triangles. The anapole order parameter $\mathbf{L}$ for this domain is also shown. Note that according to (1), $\mathbf{L}$ in general does not point along the diagonal of the rectangle.
Eqs. [9] and [10] have the usual terms proportional to $\mathbf{E} \times \mathbf{H}$, which do not contribute to the NR defined in Eq. [1]. From the cross-terms in $\mathbf{E}$ and $\mathbf{b}$ and in $\mathbf{B}$ and $\mathbf{e}$, one gets respectively Hall effects with no external $\mathbf{B}$ or no external $\mathbf{E}$: they are linear in $\mathbf{L}$ and therefore cancel in transport measurements in macroscopic samples in the sum over different domains of $\mathbf{L}$. The terms of immediate interest are proportional to

$$\mathbf{e} \times \mathbf{b} = \chi^2 (c/c^*) (\mathbf{L} \cdot (\mathbf{B} \times \mathbf{E})) \mathbf{L}. \quad (11)$$

Unlike the conventional Hall current, this contribution is "uniaxial"; it flows only along $\mathbf{L}$ and depends on the projection of $\mathbf{B} \times \mathbf{E}$ onto $\mathbf{L}$. This cannot obey Onsager reciprocity. The maximum NR is for the electric field applied parallel or perpendicular to $\mathbf{L}$, where the anomalous contribution is zero for the latter case.

On using (8) and (5) give contributions,

$$\sigma_{ab} = \sigma^H \chi^2 (c/c^*) L_a^2$$
$$\sigma_{ba} = \sigma^H \chi^2 (c/c^*) L_b^2. \quad (12)$$

These contributions are the same for all four domains. There are similar non-reciprocal contributions in $\alpha_{ab}$ and $\alpha_{ba}$.

To calculate the value of NR, we need both the off-diagonal and the diagonal components of the transport tensors. $\sigma_{aa}/\sigma_{bb}$ is measured [3,4]. As discussed above, based on available data we assume that $\sigma_{aa}/\sigma_{bb}$ behaves similarly. Thus,

$$\frac{\sigma_{aa}}{\sigma_{bb}} \approx \frac{\alpha_{aa}}{\alpha_{bb}} = r, \quad (13)$$

with a nearly temperature independent $r$ in the range of interest. The measured value [3,4] is about 1/2.

Then for $T$ just below $T^*(x)$ where $(c/c^*) \chi^2 L^2 \ll 1$

$$X(T) \propto (c/c^*) \chi^2 (L_a^2 - L_b^2) \quad (14)$$

This agrees with the experimental result that $X(T) \approx (T^*(x) - T)$, in the mean-field range in which $L^2 \propto (T^*(x) - T)$. At low temperatures, where $L^2$ is a constant, $X(T)$ is also a constant. This is also consistent with the experiments. The sign of the effect is also consistent if $L_b^2 > L_a^2$, as is expected.

Given the available data, it is impossible to calculate the magnitude of the effect and compare with the experiments. The problem is (1) that since $r$ is neither too small compared to 1 or too close to 1, the expressions for $X(T)$ cannot be simply disentangled in terms of $L$ and $r$ without separately knowing the magnitudes of $\alpha_{ab}$ and $\sigma_{ab}$; for at least some temperature, and (2) $r$ has a contribution due to chains which cannot be disentangled in the experiment so that a contribution to the magnitude of the Nernst coefficients from the planes can be separately obtained. It would be far more satisfactory to directly measure the ratio of the off-diagonal parts, $\sigma_{ab}/\sigma_{ba}$ and $\alpha_{ab}/\alpha_{ba}$ to test for violation of Onsager reciprocity directly, unlike the Nernst coefficients which has diagonal and off-diagonal parts of the two different transport tensors mixed in. We urge such experiments. Especially interesting would be the angular dependence suggested through Eq. [11].

To summarize, the experimental results on the anisotropy of the Nernst coefficient are consistent with a violation of the Onsager Reciprocity for $T < T^*(x)$ if, as reported, the diagonal components of the conductivity and as is probable, the diagonal components of the thermopower are smooth across $T^*(x)$. Such a violation is consistent with the magneto-electric symmetry already deduced in microscopic experiments in Cuprates.

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