On T-Violation Signals Originating from Magnetic Orders through Double Exchange Mechanism

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Abstract
The photon self energy tensor in a crystal with double exchange and ferromagnetic or canting magnetic orders is analyzed at zero temperature with emphasis on the signal of the time-reversal invariance breaking. Three comparable contributions, spin susceptibility, local magnetic field and spin-orbit coupling are estimated and the prospect of their experimental detection is discussed.

1. Introduction

Recently, there are growing interests in the double exchange mechanism proposed by Zener [1], Anderson and Hasegawa [2] over forty years ago. It stems from the possible relevance of the mechanism to the colossal magnetoresistance of the single perovskite material \( La_{2-x}Ca_xMnO_3 \) [3] and is conjectured also to be responsible to the newly synthesised double perovskite material \((Sr, Ba)_2YRu_{1-x}Cu_xO_6\) [4]. The physical picture of the double exchange mechanism in the latter system lies in the outer electronic configuration of the Ru-ion. Prior to doping, the Ru-ion carries a charge +5 and its outer electronic configuration is \(4d^3\), the spin of each electron are forced parallel by strong Hund’s rule coupling and the three electrons fills
in a closed multiplet of the cubic group. The total spin of each $Ru^{+5}$ is $3/2$ and interacts with each other through an antiferromagnetic super-exchange. The parent compound is therefore an antiferromagnetic insulator. When the charge balance is offset by doping of lower valence transition elements, $Cu$, holes are introduced to the outer electronic configuration and they start to hop. The system becomes a conductor and may even be a superconductor below certain temperature. The spins of these holes couple strongly with the $Ru^{+5}$ ions via Hund’s rule and bring in ferromagnetic couplings between ion spins. The prototype Hamiltonian of this mechanism reads

$$H = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j - \mu_c \sum_j \vec{B}_j \cdot \vec{S}_j$$

$$-\frac{1}{2} \sum_{ij} t_{ij} (\Psi_i^\dagger \Psi_j + \Psi_j^\dagger \Psi_i) + \frac{1}{2} \lambda \sum_j \vec{S}_j \Psi_j^\dagger \vec{\sigma} \Psi_j + H_{Coul.},$$

(1.1)

where $\vec{S}_j$ is the total spin operator of the ion core, $\Psi_j, \Psi_j^\dagger$ are the creation and the annihilation operators of the itinerant carriers, each of two components with spin up and spin down, $J_{ij}$ denotes the antiferromagnetic exchange, $t_{ij}$ denotes the hopping amplitude of the itinerant electrons and $\lambda$ denotes the Hund’s rule coupling of the itinerant electrons with the ion cores. The Coulomb interaction, $H_{Coul.}$, is given by

$$H_{Coul.} = \frac{e^2}{8\pi \varepsilon} \left( U \sum_{j,\sigma,\sigma'} : \Psi_j^\dagger \Psi_j \Psi_j^\dagger \Psi_j : + \sum_{i\neq j,\sigma,\sigma'} : \Psi_i^\dagger \Psi_i \Psi_j^\dagger \Psi_j : \right)$$

with the first term the on-site Coulomb repulsion and $\varepsilon$ the dielectric constant produced by the ion cores.

The competition between the antiferromagnetic exchange and the ferromagnetic coupling may lead to a canting magnetic order below certain temperature, as was analyzed by de-Gennes [5] and supported by specific heat measurement of the double perovskite materials [6]. In the previous paper [7], hereby referred to as paper I, we studied the magnetic ordering of the Hamiltonian (1.1). Spinwave spectrum was calculated from the Hamiltonian (1.1) with a canting magnetic order. The important role played by long range Coulomb interaction in maintaining such an order is emphasized. We also performed extensive Monte Carlo simulations on de-Gennese’s effective magnetic Hamiltonian and computed the specific heat as a function of temperature. All our results were found consistent with the experimental measurements of the specific heat and the magnetic susceptibility.

In this paper, we shall address to the optical properties of the unusual magnetic ordering caused by the double exchange. The characteristic term
of the Hamiltonian (1.1) is the Hund’s rule coupling term. A reasonable estimate \[2\] gives rise to \(\lambda S \sim 1\text{eV}\). In the paramagnetic phase, such a term causes strong scattering of itinerant holes. In the ordered phase with ferromagnetic order or canting, such a term represents an effective homogeneous magnetic field acting on the hole spins only. With the magnetic moment of a free electron, \(2\mu_{\text{Bohr}}\), the equivalent magnetic field strength is as high as

\[B_{\text{equiv}} \sim 10^4\text{Tesla},\]  \(1.3\)

(\(2\mu_{\text{Bohr}}B_{\text{equiv}} \sim \lambda S\)). Detectable effect signaling the violation of time reversal invariance is expected and such effects are the main theme of this work.

In the following two sections, we shall couple the system described by the model Hamiltonian (1.1) with radiation field calculate the photon self energy function of the medium. A f.c.c cubic structure like the double perovskite materials and and a tetragonal layer type magnetic ordering are assumed for concreteness. In contrast to the ordinary magnetooptical effect, the equivalent magnetic field (1.3), essentially rooted in the Coulomb forces, does not acts on the orbital motion directly. The optical activity comes from three different sources: 1) the Pauli term; 2) the secondary magnetooptical effect because of the real magnetic field produced by ordered spins; 3) a spin-orbital coupling which comes together with the Pauli-term in the nonrelativistic approximation of the Dirac electrons. The prospect of the experimental detection, the relevance of possible genuine superconductivity order and the validity of various approximation we made are discussed in the final section.

2. The Electromagnetic Coupling and Its Parabolic Approximation

The Hamiltonian combining the antiferromagnetic exchange, the double exchange and the electromagnetic coupling reads

\[H = \int d^3 r \frac{1}{2}(\vec{E}_{\text{tr}}^2 + \vec{B}^2) + \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j - \mu_i \sum_j \vec{B}_j \cdot \vec{S}_j + \frac{1}{2} \sum_{ij} t_{ij}(\Psi_i^\dagger U_{ij} \Psi_j + \Psi_j^\dagger U_{ji} \Psi_i) - \frac{1}{2} \lambda \sum_j \vec{S}_j \Psi_j^\dagger \vec{\sigma} \Psi_j - \mu_h \sum_j \Psi_j^\dagger \vec{\sigma} \cdot \vec{B}_j \Psi_j + H_{\text{Coul.}},\]  \(2.1\)

The Coulomb gauge is adapted with \(\vec{\nabla} \cdot \vec{A} = 0\). The transverse electric field \(\vec{E}_{\text{tr}} = -\frac{\partial \vec{A}}{\partial t}\) and the magnetic field \(\vec{B} = \vec{\nabla} \times \vec{A}\) where \(\vec{B}_j = \vec{B}(\vec{R}_j)\) with \(\vec{R}_j\) the position vector of the \(j\)th site. The lattice gauge coupling to the carrier
field is given by

\[ U_{ij} = \exp \left[ -ie \int_0^1 ds \vec{l}_{ij} \cdot \vec{A} (\vec{R}_i + s \vec{l}_{ij}) \right] \]  

(2.2)

where \( \vec{l}_{ij} \) denotes the displacement vector from the \( i \)th site to the \( j \)th site. In what follows, we shall restrict the hopping amplitude within the nearest bond only.

Following the conventions in I, we introduce the lattice frame \((x, y, z)\) with \( \hat{x}, \hat{y} \) and \( \hat{z} \) parallel to the \((100), (010)\) and \((001)\) directions of the crystal respectively. The locations of magnetic ions (total number = \( N \)) are given by

\[ \vec{R} = n_1 \vec{e}_1 + n_2 \vec{e}_2 + n_3 \vec{e}_3, \]  

(2.3)

where \( n_1, n_2 \) and \( n_3 \) are integers,

\[ \vec{e}_1 = \frac{l}{2} (\hat{y} + \hat{z}), \]  

(2.4)

\[ \vec{e}_2 = \frac{l}{2} (\hat{z} + \hat{x}), \]  

(2.5)

\[ \vec{e}_3 = \frac{l}{2} (\hat{x} + \hat{y}), \]  

(2.6)

with \( l \) the side length of a basic cube \((l = 8.16 \text{Å}, \text{for} \ Sr_2YRu_{1-x}Cu_xO_6)\).

Adapting the tetragonal layer magnetic ordering, we divide the lattice into two sublattices shown in Fig 1. with \( n_1 + n_2 = \text{even} \) on the sublattice \( A \) and \( n_1 + n_2 = \text{odd} \) on the sublattice \( B \). The expectation value of the ion spins of the sublattice \( A \) is denoted by \( \vec{S}_A = S \vec{\zeta}_A \) and that of the sublattice \( B \) by \( \vec{S}_B = S \vec{\zeta}_B \). We choose the \( \zeta \)-axis along the direction of \( \vec{S}_A + \vec{S}_B \) and the \( \eta \)-axis as

\[ \hat{\eta} = \frac{\vec{\zeta}_A \times \vec{\zeta}_B}{\sin \Theta} \]  

(2.7)

with \( \Theta \) the mutual angle between \( \vec{S}_A \) and \( \vec{S}_B \). We introduce further

\[ \vec{\xi} = \hat{\eta} \times \vec{\zeta}, \]  

(2.8)

The set \((\vec{\xi}, \hat{\eta}, \vec{\zeta})\) forms a right hand coordinate system and will be referred to as the spin frame. Without the magnetic anisotropic energy, the orientation of the spin frames does not couple with the lattice frame.

With large spin approximation, the Hamiltonian (1.1) can be written as

\[ H = H_{\text{e.m.}} + H_{\text{s.w.}} + H_{\text{Coul.}}, \]  

(2.9)
where $H_{\text{e.m.}}$ is the part of the Hamiltonian we are concerned with. Its is given by

$$H_{\text{e.m.}} = \int d^3r \frac{1}{2}(E_{ir}^2 + \vec{B}^2) - \frac{1}{2} t \sum_{<ab>} (\Psi_{a}^\dagger U_{ab} \Psi_{b} + \Psi_{b}^\dagger U_{ba} \Psi_{a})$$

$$- \frac{1}{2} t' \left[ \sum_{<aa'}} (\Psi_{a}^\dagger U_{a'a'} \Psi_{a'} + \Psi_{a'}^\dagger U_{a'a} \Psi_{a}) + \sum_{<bb'}} (\Psi_{b}^\dagger U_{bb'} \Psi_{b'} + \Psi_{b'}^\dagger U_{b'b} \Psi_{b}) \right]$$

$$- \frac{1}{2} \lambda_s \left( \sum_{a} \zeta_A \Psi_{a}^\dagger \vec{\sigma} \Psi_{a} + \sum_{b} \zeta_B \Psi_{b}^\dagger \vec{\sigma} \Psi_{b} - \mu_h \left( \sum_{a} \Psi_{a}^\dagger \vec{\sigma} \cdot \vec{B}_a \Psi_{a} + \sum_{b} \Psi_{b}^\dagger \vec{\sigma} \cdot \vec{B}_b \Psi_{b} \right) \right)$$

$$- \frac{1}{2} \mu_h \left( \sum_{a} \zeta_A \Psi_{a}^\dagger \vec{\sigma} \Psi_{a} + \sum_{b} \zeta_B \Psi_{b}^\dagger \vec{\sigma} \Psi_{b} - \mu_h \left( \sum_{a} \Psi_{a}^\dagger \vec{\sigma} \cdot \vec{B}_a \Psi_{a} + \sum_{b} \Psi_{b}^\dagger \vec{\sigma} \cdot \vec{B}_b \Psi_{b} \right) \right),$$

where $t$ and $t'$ denote the interlayer and intralayer hoppings respectively. The terms containing spin wave operators are included in $H_{\text{e.m.}}$ and have been dealt with in I.

Within Heisenberg representation, the time reversal operator $T$ transforms

$$T \vec{A}(\vec{r},t) T^{-1} = -\vec{A}(\vec{r},-t),$$

$$T \Psi_j(t) T^{-1} = \kappa \sigma_2 \Psi_j(-t)$$

and

$$T \Psi_j^\dagger(t) T^{-1} = \kappa^* \Psi_j^\dagger(-t) \sigma_2$$

with $\kappa$ a phase factor. It is easy to see that

$$TH_{\text{e.m.}} T^{-1} \neq H_{\text{e.m.}},$$

Figure 1 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 2 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 3 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 4 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 5 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 6 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 7 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 8 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 9 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$. 

Figure 10 (a) The magnetic ordering. The magnetic sites of the sublattices A and B are marked explicitly. (b) The spin frame. The angle between $\zeta_A$, and $\zeta_B$ axes is $\Theta$.
because of the terms containing Pauli matrices and therefore $H_{e.m.}$ is not invariant under a time reversal.

We may also introduce a pseudo time reversal operator $\mathcal{T}$, whose representation is
\[
\mathcal{T} \vec{A}(\vec{r}, t) \mathcal{T}^{-1} = -\vec{A}(\vec{r}, -t),
\]
(2.15)
and
\[
\mathcal{T}\Psi_j(t) \mathcal{T}^{-1} = \kappa' \Psi_j(-t),
\]
(2.16)
with $\kappa'$ another phase factor. It is interesting to notice that except the last two terms of (2.9), the rest of $H_{e.m.}$ is invariant under $\mathcal{T}$. This property is very useful for the one-loop calculations in the next section. The Hamiltonian (2.1) is also invariant under the space inversion
\[
P\vec{A}(\vec{r}, t) P^\dagger = -\vec{A}(-\vec{r}, t)
\]
(2.18)
and
\[
P\Psi_j(t) P^\dagger = \eta \Psi_j^*(t)
\]
(2.19)
with $j^*$ the inversion image of $j$ and $\eta$ a phase factor.

The complicated dependence of $U_{ij}$ makes the perturbative expansion in terms of the fine structure constant rather cumbersome. we shall make a parabolic approximation for the rest of the paper. We found in I that, at the absence of the vector potential, (2.10) reduces to $\sum_{\vec{p}} \Psi_\vec{p}^\dagger E_\vec{p} \Psi_\vec{p}$ with $4 \times 1$ column matrix $\Psi_\vec{p}$ and $E_\vec{p}$ the $4 \times 4$ hermitian matrix.

\[
E_\vec{p} = -t'u_{\vec{p}} - tv_{\vec{p}}\rho_1 - \frac{1}{2} \lambda S(\rho_+ \hat{\zeta}_A \cdot \vec{\tau} + \rho_- \hat{\zeta}_B \cdot \vec{\tau}),
\]
(2.20)
where
\[
\tau_1 = \begin{pmatrix} \sigma_1 & 0 \\ 0 & \sigma_1 \end{pmatrix}, \tau_2 = \begin{pmatrix} \sigma_2 & 0 \\ 0 & \sigma_2 \end{pmatrix}, \tau_3 = \begin{pmatrix} \sigma_3 & 0 \\ 0 & \sigma_3 \end{pmatrix}
\]
(2.21)
and
\[
\rho_1 = \begin{pmatrix} 0 & I \\ I & 0 \end{pmatrix}, \rho_2 = \begin{pmatrix} 0 & -iI \\ iI & 0 \end{pmatrix}, \rho_3 = \begin{pmatrix} I & 0 \\ 0 & -I \end{pmatrix}
\]
(2.22)
with
\[
[\rho_a, \tau_b] = 0.
\]

The kinetic energy, $\sum_{\vec{p}} \Psi_\vec{p}^\dagger E_\vec{p} \Psi_\vec{p}$, is invariant under the $C$-transformation, $C\Psi_\vec{p} C^{-1} = -\rho_1 \tau_3 \Psi_\vec{p}$ and $C^2 = 1$. This symmetry can be diagonalized through the unitary transformation $\Psi_\vec{p} = V \psi_\vec{p}$ with
\[
V = \frac{1}{\sqrt{2}} (\rho_- + \rho_+ \tau_3)(\rho_1 + \rho_3) \left( \cos \frac{\Theta}{4} - i \tau_2 \sin \frac{\Theta}{4} \right).
\]
(2.24)
We find that
\[ V^\dagger E_P V = -t' u_{\vec{p}} - tv_{\vec{p}} \rho_3 \left( \tau_3 \cos \frac{\Theta}{2} - \tau_1 \sin \frac{\Theta}{2} \right) - \frac{1}{2} \lambda S \tau_3, \]  
which is easily diagonalized and produces four energy bands:
\[ \epsilon_{\vec{p}} = -t'(u_{\vec{p}} + \mu) \pm \Delta_{\vec{p}}^\pm \]  
with
\[ \Delta_{\vec{p}}^\pm = \sqrt{\frac{1}{4} \lambda^2 S^2 \pm \lambda Stv_{\vec{p}} \cos \frac{\Theta}{2} + t^2 v_{\vec{p}}^2}, \]
where \( \pm \) of \( \Delta_{\vec{p}} \) labels the eigenvalue of the \( C \) operator. The lowest band corresponds to
\[ \epsilon_{\vec{p}} = -t'(u_{\vec{p}} + \mu) - \Delta_{\vec{p}}^+ \]
which overlaps partially at the top with the next lowest band, i.e., \( \epsilon_{\vec{p}} = -t'(u_{\vec{p}} + \mu) - \Delta_{\vec{p}}^- \). In what follows we shall assume that only the band \( (2.28) \) is partially filled, a likely situation with doping conditions of \((Sr, Ba)_2YRu_{1-x}Cu_xO_6\).
With the parabolic approximation we have
\[ u_{\vec{p}} = 2 - \frac{l^2}{4} (p_x^2 + p_y^2) \]  
and
\[ v_{\vec{p}} = 4 - \frac{l^2}{4} (p_x^2 + p_y^2 + 2p_z^2). \]
Regarding \( \psi_{\vec{p}} \) as the Fourier component of a \( 4 \times 1 \) fermion field \( \psi(\vec{r}) \) in continuum, we have \( \frac{2}{N} \sum_{\vec{p}} = \int \frac{d^3 \vec{p}}{(2\pi)^3} \) and the "coordinate" representation of kinetic energy operator, \((2.25)\), reads
\[ K = \int d^3 \vec{r} \left[ \frac{1}{2} \vec{v}_\perp \psi^\dagger \cdot \left( \frac{1}{m'} + \frac{1}{m} \rho_3 \tau_3^t \right) \vec{v}_\perp \psi + \frac{1}{m} \partial_z \psi^\dagger \rho_3 \tau_3^t \partial_z \psi \right. \]
\[ - \nu \psi^\dagger \rho_3 \tau_3^t \psi - \mu_c \psi^\dagger \psi - \frac{1}{2} \lambda S \psi^\dagger \tau_3 \psi \],
where the parameters \( m, m' \) and \( \nu \) are related to the discrete Hamiltonian through \( m'^{-1} = tl^2/2, m^{-1} = tl^2/2, \nu = 4t, \mu_c = \mu + 2t' \). Replace the ordinary derivatives by the covariant ones, we obtain the continuum approximation of \((2.10)\), i.e.
\[ H_{e.m.} = \int d^3 \vec{r} \left[ \frac{1}{2} (\vec{E}^2_{ir.} + \vec{B}^2) + \int d^3 \vec{r} \left[ \frac{1}{2} (\vec{v}_\perp + ieA_\perp) \psi^\dagger \cdot \left( \frac{1}{m'} + \frac{1}{m} \rho_3 \tau_3^t \right) (\vec{v}_\perp - ieA_\perp) \psi \right] \right] \]
\[
+ \frac{1}{m} \left( \frac{\partial}{\partial z} + ieA_z \right) \psi^\dagger \rho_3 \tau'_3 \left( \frac{\partial}{\partial z} - ieA_z \right) \psi \\
- \nu \psi^\dagger \rho_3 \tau'_3 \psi - \mu_c \psi^\dagger \psi - \frac{1}{2} \lambda S \psi^\dagger \tau_3 \psi - \frac{g_e}{2} \mu_B \psi^\dagger \bar{\alpha} \cdot \vec{B} \psi \right], \tag{2.32}
\]

where
\[
\bar{\alpha} = V^\dagger \vec{\tau} V = -\rho_1 \tau'_1 \hat{\xi} - \rho_1 \tau_2 \hat{\eta} + \tau_3 \hat{\zeta}, \tag{2.33}
\]
\[
\tau'_3 = \tau_3 \cos \frac{\Theta}{2} - \tau_1 \sin \frac{\Theta}{2}, \tag{2.34}
\]
\[
\tau'_1 = \tau_3 \sin \frac{\Theta}{2} + \tau_1 \cos \frac{\Theta}{2}, \tag{2.35}
\]
e is positive for holes and negative for electrons and we have substituted \(g_e \nu_B\) for \(\mu_h\).

It is well known that, the Hamiltonian (2.10) and (2.32) produce the same physics as long as the length scales involved are much larger than the lattice spacing, \(l\). The Hamiltonian (2.32) is not bounded from below but this does no harm for the perturbative calculations provide the condition of the parabolic approximation is met. Like the Hamiltonian (2.10) the Hamiltonian (2.32) is not invariant under a time reversal. But the pseudo time reversal transformation leaves all terms except the last one (Pauli term) invariant.

The electric current operator of the system consists of three terms
\[
\vec{J}(\vec{r}, t) = \vec{J}_d(\vec{r}, t) + \vec{J}_p(\vec{r}, t) + \vec{J}_s(\vec{r}, t) \tag{2.36}
\]

where \(\vec{J}_d\), \(\vec{J}_p\) and \(\vec{J}_s\) denote the diamagnetic, paramagnetic and spin part and their expressions are listed below
\[
\vec{J}_d(\vec{r}, t) = -\frac{e^2}{2} \psi^\dagger \left( \frac{1}{m} + \frac{1}{m} \rho_3 \tau'_3 \right) \psi \vec{A} - \frac{e^2}{m} \psi^\dagger \rho_3 \tau'_3 \psi \vec{A}_z \hat{z}; \tag{2.37}
\]
\[
\vec{J}_p(\vec{r}, t) = -\frac{ie}{2} \left[ \psi^\dagger \left( \frac{1}{m} + \frac{1}{m} \rho_3 \tau'_3 \right) \vec{\nabla} \psi - \text{h.c.} \right] - \frac{ie}{m} \left( \psi^\dagger \rho_3 \tau'_3 \frac{\partial}{\partial z} \psi - \text{h.c.} \right) \hat{z} \tag{2.38}
\]
and
\[
\vec{J}_s(\vec{r}, t) = \frac{g_e}{2} \mu_B \vec{\nabla} \times \psi^\dagger \bar{\alpha} \psi. \tag{2.39}
\]
3. The Response Functions

When the system is illuminated by a beam of light with a wave vector $\vec{k}$. The optical response is entirely determined by the transverse components of the retarded photon self energy tensor in the light frame, i.e.

$$\sigma_{\alpha\beta}(\omega) = (\hat{e}_\alpha)_i \Pi_R^{ij}(\omega, \vec{k})(\hat{e}_\beta)_j$$ (3.1)

where, at zero temperature,

$$\text{Re}\Pi_R^{ij}(\omega, \vec{k}) = \text{Re}\Pi_{ij}(\omega, \vec{k})$$ (3.2)

and

$$\text{Im}\Pi_R^{ij}(\omega, \vec{k}) = \text{sign}(\omega)\text{Im}\Pi_{ij}(\omega, \vec{k})$$ (3.3)

with $\Pi(\omega, \vec{k})$ time-ordered self energy tensor which can be calculated diagrammatically. For a system invariant under the space inversion, we expect

$$\Pi_{ij}(\omega, \vec{k}) = \Pi_{ij}(\omega, -\vec{k}).$$ (3.4)

For a system invariant under time reversal, we have

$$\Pi_{ij}(\omega, \vec{k}) = \Pi_{ji}(\omega, -\vec{k}).$$ (3.5)

Come to the system we are considering, the time reversal invariance is spontaneously broken by the magnetic ordering but inversion symmetry is intact. The self energy tensor will acquire an antisymmetric part, i.e.

$$\Pi_{ij}(\omega, \vec{k}) - \Pi_{ji}(\omega, \vec{k}) \neq 0,$$ (3.6)

which we shall focus our attention on for the rest of the article.

The one-loop diagrams for $\Pi_{ij}(\omega, \vec{k})$ are displayed in Fig. 2.

The evaluation of them is straightforward and we tabulate below the results under the approximation $\lambda S >> t, t' \omega \sim \lambda S$ and $\rho_h l^3 << 1$ where $\rho_h$ is the number of itinerant holes per unit volume,

$$\rho_h = \int \frac{d^3 \vec{p}}{(2\pi)^3} \theta(\mu_c - \epsilon_\vec{p})$$ (3.7)
with \( \theta(x) = \frac{1}{2}(1 + \text{sign}(x)) \). We divide \( \Pi_{ij}(\omega, \vec{k}) \) into three terms,

\[
\Pi_{ij}(\omega, \vec{k}) = \Pi_{ij}^{(1)}(\omega, \vec{k}) + \Pi_{ij}^{(2)}(\omega, \vec{k}) + \Pi_{ij}^{(3)}(\omega, \vec{k})
\]  

(3.8)

The \( \Pi^{(1)} \) term comes from the orbital motion only and is given by

\[
\Pi_{ij}^{(1)}(\omega, \vec{k}) = e^2 \rho_h \left[ \frac{1}{m'} (\delta_{ij} - \delta_{iz}\delta_{jz}) + \frac{1}{m} \cos \frac{\Theta}{2} (\delta_{ij} + \delta_{iz}\delta_{jz}) \right]
\]

\[
- i \int d^3 \vec{r} e^{-i\vec{k} \cdot \vec{r}} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle |TJ_{p,i}(\vec{r}, t)J_{p,j}(0, 0)| \rangle >
\]

\[
= e^2 \rho_h \left[ \frac{1}{m'} (\delta_{ij} - \delta_{iz}\delta_{jz}) + \frac{1}{m} \cos \frac{\Theta}{2} (\delta_{ij} + \delta_{iz}\delta_{jz}) \right]
\]

\[
- e^2 \rho_h \frac{\lambda S}{m^2 \lambda^2 S^2 - \omega^2} \left[ <p_{\perp}^2 > (\delta_{ij} - \delta_{iz}\delta_{jz}) + 8 <p_{z}^2 > \delta_{iz}\delta_{jz} \right] \sin^2 \frac{\Theta}{2},
\]  

(3.9)

where

\[
<F> \equiv \frac{1}{\rho_h} \int \frac{d^3 \vec{p}}{(2\pi)^3} \theta(-\varepsilon_{\vec{p}}) F(\vec{p}).
\]  

(3.10)

The \( \Pi^{(2)} \) term of (3.8) represent the cross term between the paramagnetic current and the spin current and is given by

\[
\Pi_{ij}^{(2)}(\omega, \vec{k}) = - i \int d^3 \vec{r} e^{-i\vec{k} \cdot \vec{r}} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle |T|J_{s,i}(\vec{r}, t)J_{s,j}(0, 0)+J_{s,i}(\vec{r}, t)J_{p,j}(0, 0)| \rangle >
\]

\[
= i[\epsilon_{imn} k_m \hat{\zeta}_n \Lambda_j(\omega, \vec{k}) - \epsilon_{jmn} k_m \hat{\zeta}_n \Lambda_i(\omega, \vec{k})]
\]  

(3.11)

where

\[
\Lambda_j(\omega, \vec{k}) = g_e \frac{\mu_B \rho_h}{2m\omega} \left[ \left( \frac{m}{m'} \cos \frac{\Theta}{2} + \frac{\lambda^2 S^2 \cos^2 \frac{\Theta}{2} - \omega^2}{\lambda^2 S^2 - \omega^2} \right) k_j(1 - \delta_{jz}) \right.
\]

\[
+ \left. 2 \frac{\lambda^2 S^2 \cos^2 \frac{\Theta}{2} - \omega^2}{\lambda^2 S^2 - \omega^2} k_z \delta_{jz} \right].
\]  

(3.12)

Finally, the \( \Pi^{(3)} \) term is related to the spin susceptibility tensor \( \chi_{ij}(\omega) \) via

\[
\Pi_{ij}^{(3)}(\omega, \vec{k}) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle |TJ_{s,i}(\vec{r}, t)J_{s,j}(0, 0)| \rangle = \epsilon_{imn} \epsilon_{jm'n'} k_m k_{m'} \chi_{mn'}(\omega),
\]  

where

\[
\chi_{ij}(\omega) = R_{i\mu} R_{j\nu} \chi_{\mu\nu}(\omega)
\]  

(3.13)

(3.14)
with the indices $\mu, \nu$ referring to axes of the spin frame ($\mu, \nu = \xi, \eta, \zeta$) and $R_{\mu\nu}$ the elements of the orthogonal matrix

$$
R = \begin{pmatrix}
\hat{x} \cdot \hat{\xi} & \hat{x} \cdot \hat{\eta} & \hat{x} \cdot \hat{\zeta} \\
\hat{y} \cdot \hat{\xi} & \hat{y} \cdot \hat{\eta} & \hat{y} \cdot \hat{\zeta} \\
\hat{z} \cdot \hat{\xi} & \hat{z} \cdot \hat{\eta} & \hat{z} \cdot \hat{\zeta}
\end{pmatrix}.
$$

The nonvanishing components of $\chi_{\alpha\beta}(\omega)$ are

$$
\chi_{\xi\xi}(\omega) = -\frac{1}{2} g_e^2 \rho_B \mu_B^2 \frac{\lambda S \cos^2 \frac{\Theta}{2}}{\lambda^2 S^2 - \omega^2},
$$

$$
\chi_{\eta\eta}(\omega) = -\frac{1}{2} g_e^2 \rho_B \mu_B^2 \frac{\lambda S}{\lambda^2 S^2 - \omega^2},
$$

$$
\chi_{\xi\eta}(\omega) = -\chi_{\eta\xi}(\omega) = -\frac{i}{2} g_e^2 \rho_B \mu_B^2 \frac{\omega \cos \frac{\Theta}{2}}{\lambda^2 S^2 - \omega^2},
$$

$$
\chi_{\zeta\zeta}(\omega) = -\frac{1}{2} g_e^2 \rho_B \mu_B^2 \frac{\lambda S \sin^2 \frac{\Theta}{2}}{\lambda^2 S^2 - \omega^2}.
$$

We observe that 1) The antisymmetric part comes only from $\Pi^{(2)}$ and $\Pi^{(3)}$ terms (the statement is true without the approximation $\lambda S >> t, t'$ and $n_h l^3 << 1$), while $\Pi^{(1)}$ remains symmetric, a consequence of $T$-invariance. 2) With the large double exchange approximation, the symmetric part of $\Pi_{ij}(\omega, \vec{k})$ is dominated by the first term of (3.9) and the antisymmetric terms are smaller than this term by a factor of the order of $k^2/m\lambda S$. For $\omega$ not at the pole, $\pm \lambda S$, we have $\Pi^{R}_{ij}(\omega, \vec{k}) = \Pi_{ij}(\omega, \vec{k})$ and from this the expression of $\sigma_{\alpha\beta}(\omega, \vec{k})$ follows.

The propagation mode of a photon is determined by

$$
[(\omega^2 - k^2)\delta_{\alpha\beta} - \sigma_{\alpha\beta}(\omega, \vec{k})] \phi_\beta = 0
$$

with $\vec{\phi}$ the polarization vector, $\vec{\phi} \perp \vec{k}$. The $T$-violation implies that the eigenmodes are elliptically polarized instead of linearly polarized and the system possesses the optical activity.

Consider a special circumstance, in which $\vec{k}, \hat{\zeta}$ and $\hat{z}$ are all parallel, and the magnetic order is purely ferromagnetic. The self energy tensor $\sigma_{\alpha\beta}$ reduces to

$$
\sigma_{\alpha\beta}(\omega, \vec{k}) = e^2 \rho_B \left( \frac{1}{m'} + \frac{1}{m} \right) \delta_{\alpha\beta} - \frac{i}{2} k^2 \epsilon_{\alpha\beta} g_e^2 \rho_B \mu_B^2 \frac{\omega}{\lambda^2 S^2 - \omega^2},
$$

(3.21)
where we have shown explicitly the symmetric and antisymmetric parts of \(\sigma_{\alpha\beta}(\omega, \vec{k})\) and only the leading order terms are retained in each part. The eigenmode equation (3.20) are reduced to

\[
\left[ (\omega^2 - \omega_p^2) \delta_{\alpha\beta} - n^2 \omega^2 \left( 1 - \frac{i}{2} \epsilon_{\alpha\beta} g e \rho_h \mu_B^2 \frac{\omega}{\lambda^2 S^2 - \omega^2} \right) \right] \phi_{\beta} = 0, \tag{3.22}
\]

where we have introduced the index of refraction, \(n(\omega)\) via \(k = n(\omega) \omega\) and the plasma frequency \(\omega_p^2 = e^2 \rho_h (\frac{1}{m} + \frac{1}{m'} \cos \Theta)\). Clearly, the eigenmodes of (3.20) are two circularly polarized photon with opposite senses. The corresponding indices of refraction differ by an amount

\[
\Delta n(\omega) = \frac{1}{2} \sqrt{\epsilon_0 g \mu_B^2 \frac{\omega}{\lambda^2 S^2 - \omega^2}} \tag{3.23}
\]

with \(\epsilon_0 = 1 - \frac{\omega_p^2}{\omega^2}\), which gives rise to a Faraday angle

\[
\Delta \theta = \pi \Delta n(\omega) \tag{3.24}
\]

per wavelength. For \(Sr_2YRu_{1-x}Cu_xO_6\), we have \(\rho_h = 2x/v\) with \(v = l^3/4 = 136 A^3\). Assuming \(g_e = 2\), \(\Theta = 0\) and \(\frac{1}{m} + \frac{1}{m'} = \frac{1}{m_e}\), we obtain:

\[
\Delta \theta = 0.62 \times 10^{-4} \frac{\sqrt{\epsilon_0 x \omega}}{\lambda S^2 - \omega^2} \tag{3.25}
\]

with \(\lambda S\), \(\omega\) in ev and \(\Delta \theta\) in radians.

There are two additional contribution to the \(T\)-violation signal which are not included in the Hamiltonian (2.32) and their magnitudes turn out to be comparable to (3.25).

1. The secondary magnetooptical effect The aligned ion-spins will produced a net magnetic field, which gives rise to the ordinary magnetooptical effect [8]. For a magnetic field \(\vec{B} = B \hat{z}\) and \(\vec{k} = k \hat{z}\), the Farady angle reads

\[
\Delta \theta' = \pi \frac{\omega_p^2 \omega_B}{\sqrt{\epsilon_0 \omega^3}} \tag{3.26}
\]

with \(\omega_B = eB (\frac{1}{m} + \frac{1}{m'} \cos \Theta)\) the cyclotron frequency (See appendix A for the derivation with a general anisotropy). For a spheroidal sample with \(\vec{M}\) parallel to its symmetry axis, the average magnetic field is

\[
\vec{B} = \gamma \vec{M}
\]

with \(\vec{M}\) the magnetization given by

\[
\vec{M} = g_i \mu_B S \frac{\cos \Theta}{2}
\]
with \( g_i \) the Lande factor of the core ion, where the contribution from the itinerant holes has been neglected on account of low doping condition. The factor \( \gamma \) changes from zero in the limit of a thin disc perpendicular to the field to one in the limit of a long rod along the field. Assuming \( g_i = 2, S = \frac{3}{2}, \Theta = 0 \) and \( \frac{1}{m} + \frac{1}{m} \sim \frac{1}{m_e} \), we find the corresponding Faraday angle

\[
\Delta \theta' = 1.89 \times 10^{-3} \frac{\gamma x}{\sqrt{\epsilon_0 \omega^3}}.
\] (3.27)

Though this effect could be considerably larger than (3.25) but the double exchange effect may still be detectable because of its resonance nature.

2. Spin-orbit coupling

It is interesting to know there is a relativistic term which turns out to be comparable with (3.25). The physical reason is the same as that for the spin-orbital coupling of atomic levels. In what follows, we give a classical argument [9] and defer the quantum mechanical treatment to the appendix B. For an electron(hole) moving in an external electromagnetic field \((\vec{E}, \vec{B})\) at velocity \(\vec{v}(v << 1)\), The spin precession equation reads

\[
\frac{d\vec{s}}{dt} = \vec{s} \times \left( \frac{e}{m_e} \vec{B}' - \vec{\omega}_T \right),
\] (3.28)

where \( \vec{B}' \) is the magnetic field in the rest frame of the electron(hole) and \( \vec{\omega}_T \) is the angular velocity of Thomas precession. We have set \( g_e = 2 \) in (3.28) since we do not know how to renormalize the bare value in a medium for this effect. The equation (3.28) can be derived with a Hamiltonian term

\[
H' = -(2e\mu_B \vec{B}' - \vec{\omega}_T) \cdot \vec{s}.
\] (3.29)

To the first power of the velocity, we have

\[
\vec{B}' = \vec{B} - \vec{v} \times \vec{E}
\] (3.30)

and

\[
\vec{\omega}_T = -\frac{1}{2} \vec{v} \times \vec{a} = -\mu_B \vec{v} \times \vec{E}
\] (3.31)

with \( \vec{a} = \frac{e}{m_e} \vec{E} \) to the order of the approximation. Substituting (3.30) and (3.31) into (3.29) and express \( \vec{v} \) in terms of the canonical momentum and the gauge potential, \( \vec{v} = \frac{1}{m_e}(\vec{p} - e\vec{A}) \), we obtain

\[
H' = -2\mu_B \vec{s} \cdot \vec{B} - \frac{e}{2m_e} \vec{s} \cdot \vec{E} \times (\vec{p} - e\vec{A}).
\] (3.32)
as an improved Pauli term. The term $\vec{E} \times \vec{A}$ pick up the information of the violation of the time reversal invariance with the corresponding Farady angle per wavelength given by

$$\Delta \theta'' = \frac{\pi e^2 \rho_0}{2\sqrt{\epsilon_0 m_e \omega}} \cos \frac{\Theta}{2} = 0.62 \times 10^{-4} \frac{x}{\sqrt{\epsilon_0 \omega}} \cos \frac{\Theta}{2}. \quad (3.33)$$

4. Discussions

In the previous sections, we analyzed the photon self energy tensor in a canting magnetic ordering, focusing the signal of the $T$-violation. There are three contributions to the optical activity, eqs. (3.25), (3.27) and (3.28). Although the equivalent magnetic field due to the double exchange is huge, the magnitude of the Farady angle turns out rather small. The reason for this is that the double exchange acts only on the carrier spins. The orbital motion, neglecting the weak magnetic field produced by the ordered ion spins, does not give rise any $T$-violation signal, on account of the $T$ invariance defined in eqs.(2.15-17). Such an small effect lies at the border of the detection precision for a sample of one wavelength thick, as inferred from the delicate circular dichroism experiment, designed to test the anyon model of the high $T_C$ superconductivity [10]. On the other hand, the Farady angle associated with $T$-violation accumulates as the photon traverses back and forth inside the sample [9]. The effect may be enhanced through multiple reflections at the inner surface of the sample.

For the double exchange energy $\lambda S$ comparable with the photon energy, $\omega$, resonance character will show up in the contribution from the spin susceptibility, (3.25). A closer look will reveal an interesting point which has been watered down in the leading order of the strong double exchange approximation made in the eqs. (3.9-19). According to the Hamiltonian (2.32), the transition involved in the photon self energy includes both $C$ preserving ones and $C$ flipping ones. In the former case, the energy denominator for the transition reads

$$2\Delta_{\vec{p}}^{\pm} \pm \omega = \lambda S \pm \omega + 2t_{\vec{p}}\vec{p} \cos \frac{\Theta}{2} + O\left(\frac{t^2}{\lambda S}\right), \quad (4.1)$$

while in the latter case, the corresponding energy denominator is

$$\Delta_{\vec{p}}^{\pm} + \Delta_{-\vec{p}}^{\pm} \pm \omega = \lambda S \pm \omega + \frac{t^2}{\lambda S} v_{\vec{p}} \cos \frac{\Theta}{2}. \quad (4.2)$$
Upon integration over $\vec{p}$ within the Fermi sea, the resonance signal will be slightly smeared. Such a smearing effect for the $\mathcal{C}$ flipping transition is of one order higher than that for the $\mathcal{C}$ preserving transition. The transition contributing to $\chi_{\xi \eta}(\omega)$ is entirely $\mathcal{C}$ flipping type and the resonance signal in the Farady angle (3.25) should be fairly sharp. This feature is also independent of the parabolic approximation we made in the calculation.

The frequency dependences of the other two contributions are rather robust. The former involves the transition among different Landau levels caused by the real local magnetic field of the ordered magnetic moments of the ion cores. This field is in general fairly weak, of the order of 1000 Gauss, the level spacing associated with this magnetic field is tiny in comparison to the photon energy, $\omega$ and the energy denominator is dominated by the photon energy. The relativistic term (3.28), however, is the consequence of the virtual transition between the Dirac sea and the Fermi sea and the corresponding energy denominator is dominated by twice of the rest energy of an electron.

Experimentally, superconductivity was also discovered the double perovskite samples. A natural question arise as to the effect of the long range order on the optical activity calculated with normal electrons. For the photon in the visible region, its energy are much higher than the gap-energy and correction to and $\Pi_{ij}^{(2)}(\omega, \vec{k})$, $\Pi_{ij}^{(3)}(\omega, \vec{k})$ is of higher orders. Only thing we need to watch out is whether there is small term coming from $\Pi_{ij}^{(1)}(\omega, \vec{k})$. The long range order can be introduced by adding a pairing term to the Hamiltonian (2.32). The total Hamiltonian reads now,

$$
\mathcal{H} = H + \sum_{\vec{q}} \beta_{\vec{q}}^\dagger \beta_{\vec{q}},
$$

(4.3)

where $a_{\vec{p}}$, $a_{\vec{p}}^\dagger$ stands for the annihilation and creation operators of the itinerant holes, and $\beta_{\vec{q}}$, $\beta_{\vec{q}}^\dagger$ stands for composite boson operators which represent Cooper pairs, i.e.,

$$
\beta_{\vec{q}} = \frac{1}{\sqrt{N}} \sum_{\vec{p}}' g_{\vec{p},\vec{q}} a_{\vec{p}} a_{\vec{p}+\vec{q}/2} - a_{\vec{p}+\vec{q}/2}
$$

(4.4)

with $\sum_{\vec{p}}'$ extends over half of the Brillouin zone only. The pairing wave function, $g_{\vec{p},\vec{q}}$, is odd under the space inversion, i.e.

$$
g_{\vec{p},\vec{q}} = -g_{-\vec{p},\vec{q}}
$$

(4.5)

on account of the anticommutation relation among $a_{\vec{p}}$’s. The superconductivity is implemented through the condensation of the pairing operator of
zero total momentum, i.e.

$$\langle S|\beta_{\vec{q}=0}|S \rangle = \sqrt{N}B,$$

(4.6)

where $B$ is the long range order parameter. The wave function of the ground state is then

$$|S\rangle = \prod_{\vec{p}}^\prime (\cos \theta_{\vec{p}} - e^{-i\gamma_{\vec{p}}} \sin \theta_{\vec{p}} a_{\vec{p}}^{\dagger} a_{-\vec{p}}^{\dagger})|0\rangle,$$

(4.7)

where $\cos 2\theta_{\vec{p}} = \frac{\epsilon_{\vec{p}}}{E_{\vec{p}}}$ and $\sin 2\theta_{\vec{p}} = \frac{|\delta_{\vec{p}}|}{E_{\vec{p}}}$ with $E_{\vec{p}} = \sqrt{\epsilon_{\vec{p}}^2 + |\delta_{\vec{p}}|^2}$ and $|\delta_{\vec{p}}|e^{i\gamma_{\vec{p}}} = Bg_{\vec{p},\vec{q}} \perp \vec{q}=0$. Evidently, both the standard time reversal $T$ and the pseudo time reversal $T$ do not leave the ground state (4.7) unchanged. On the other hand, The following antiunitary transformation

$$T' a_{\vec{p}} T'^{-1} = ie^{i\gamma_{\vec{p}}} a_{\vec{p}}$$

(4.8)

leaves both the pairing Hamiltonian as well as the ground state (4.7) invariant. This, together with the invariance under the modified space inversion in $I, P = Pe^{\pi Q}$, implies that $\Pi^{(1)}_{ij}(\omega,0) = \Pi^{(1)}_{ji}(\omega,0)$ and the T-violation signal is suppressed relative to $\Pi^{(1)}_{ij}(\omega,0)$ at least by a factor $k^2/p_F^2$ with $p_F$ the Fermi-momentum. The very fact that they must vanish as the gap energy vanishes pushes it much smaller than the magnitude of $\chi_{\xi_\eta}(\omega)$. Therefore the estimate of the T-violation signals (3.25) and (3.28) with normal electrons applies to the superconducting phase as well.

As was pointed out in [11] in the context of Raman scattering, the way we introduce the gauge invariant coupling, eqs. (2.11) and (3.32), requires that the photon frequency in the energy denominators for the transition to other bands not covered by the model Hamiltonian (1.1) can be dropped. This may be rather marginal for real system. Also the impurity effect, which may be relevant to the real system has been neglected.

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Appendix A

For the ordinary magneto-optic effect, we consider only the conduction band. The effective Hamiltonian with the parabolic approximation reads

$$H = \frac{1}{2} \int d^3\vec{r} \left[ \frac{1}{2m_1} \left( \frac{\partial}{\partial x} + ieA_x \right) \psi^\dagger \left( \frac{\partial}{\partial x} - ieA_x \right) \psi ight] + \frac{1}{2m_2} \left( \frac{\partial}{\partial y} + ieA_y \right) \psi^\dagger \left( \frac{\partial}{\partial y} - ieA_y \right) \psi + \frac{1}{2m_3} \left( \frac{\partial}{\partial z} + ieA_z \right) \psi^\dagger \left( \frac{\partial}{\partial z} - ieA_z \right) \psi \right],$$  \hspace{1cm} (A.1)

where we have introduced general anisotropy and the symmetry of (A.1) is orthohmobic. In our case, which is tetragonal

$$\frac{1}{m_1} = \frac{1}{m_2} = \frac{1}{m_l} + \frac{1}{m} \cos \Theta \frac{1}{2}$$ \hspace{1cm} (A.2)

and

$$\frac{1}{m_3} = \frac{2}{m} \cos \Theta \frac{1}{2}.$$ \hspace{1cm} (A.3)

The gauge potential $\vec{A}(\vec{r},t)$ is chosen to produce a static and homogeneous magnetic field,

$$\vec{B} = \vec{\nabla} \times \vec{A}(\vec{r},t)$$ \hspace{1cm} (A.2)

and a time dependent and homogeneous electric field,

$$\vec{E} = -\frac{\partial \vec{A}}{\partial t}.$$ \hspace{1cm} (A.3)

The electric current operator reads

$$J_a = -\frac{ie}{m_a} \psi^\dagger \left( \frac{\partial}{\partial x_a} - ieA_a \right) \psi + h.c.$$ \hspace{1cm} (A.4)

with $a = 1, 2, 3$, and the charge density operator is

$$J_0 = e \psi^\dagger \psi.$$ \hspace{1cm} (A.5)

Let $| \rangle$ denote the ground state of (A.1). It follows from the translation invariance of $\vec{E}$ and $\vec{B}$ and the gauge invariance of (A.1) that the expectation values $< | \vec{J}(\vec{r},t) | >$ and $< | J_0(\vec{r},t) | >$ are coordinate independent. Introducing the operators

$$\vec{J} = \frac{1}{\Omega} \int d^3\vec{r} \vec{J}(\vec{r},t)$$ \hspace{1cm} (A.6)
and
\[ \mathcal{J}_0 = \frac{1}{\Omega} \int d^3\vec{r} J_0(\vec{r}, t), \] (A.7)
we have
\[ \vec{j} \equiv < |\vec{J}(\vec{r}, t)| > = < |\vec{J}| > \] (A.8)
and
\[ e \rho_h \equiv < |J_0(\vec{r}, t)| > = < |\mathcal{J}_0| > \] (A.9)
with \( \rho_h \) the number density of the carriers (holes in our case). In Heisenberg representation, the operator equation of motion for \( \vec{J} \) is identical in form to the classical one, i.e.
\[ \frac{d\vec{J}_a}{dt} = e^2 \frac{\mathcal{J}_0 E_a}{m_a} + e \sum_{b,c} \epsilon_{abc} \mathcal{J}_b B_c \] (A.10)
and \( \frac{d}{dt} \mathcal{J}_0 = 0 \) on account of the charge conservation. Taking the expectation value of (A.10) with respect to \( |> \), we find that
\[ \frac{dj_a}{dt} = e^2 \rho_h \frac{\mathcal{J}_0 E_a}{m_a} + e \sum_{b,c} \epsilon_{abc} J_b B_c. \] (A.11)

The anisotropy can be scaled away as:
\[ j_a = \frac{1}{\sqrt{m_a}} \hat{j}_a, \] (A.12)
\[ E_a = \sqrt{m_a} \hat{E}_a \] (A.13)
and
\[ B_c = \sqrt{m_a m_b} \hat{B}_c \] (A.14)
with \( a, b, c \) a permutation of 1, 2, 3. Then
\[ \frac{d\hat{j}_a}{dt} = e^2 \rho_h \hat{E}_a + e \sum_{b,c} \epsilon_{abc} \hat{J}_b \hat{B}_c. \] (A.15)
For \( \hat{E}_a = \hat{E}_a(\omega)e^{-i\omega t} \), we find \( \hat{j}_a = \hat{j}_a(\omega)e^{-i\omega t} \) with
\[ \hat{j}_a(\omega) = \sum_b \hat{\sigma}_{ab}(\omega) \hat{E}_b(\omega), \] (A.16)
where
\[
\hat{\sigma}_{ab}(\omega) = i \frac{e^2 \rho_h \omega}{\omega^2 - \omega_B^2} \left( \delta_{ab} - \frac{e^2 \rho_h}{\omega^2} \hat{B}_a \hat{B}_b \right) - \frac{e^3 \rho_h}{\omega^2 - \omega_B^2} \sum_c \epsilon_{abc} \hat{B}_c
\]  
(A.17)

with \(\omega_B = e \sqrt{\frac{B_1^2}{m_2 m_3} + \frac{B_2^2}{m_3 m_1} + \frac{B_3^2}{m_1 m_2}}\) the cyclotron frequency. Undoing the scale transformations (A.12-14), we obtain the complex conductivity tensor, \(\sigma_{ab}(\omega) = \frac{1}{\sqrt{m_a m_b}} \hat{\sigma}_{ab}(\omega)\). Substituting into the standard formula, we end up with the dielectric tensor with respect to the lattice frame
\[
\varepsilon_{ab}(\omega) = \delta_{ab} - \frac{e^2 \rho_h}{\sqrt{m_a m_b} (\omega^2 - \omega_B^2)} \left( \delta_{ab} - \frac{e^2 \rho_h}{\omega^2} \hat{B}_a \hat{B}_b + i \frac{e}{\sqrt{m_a m_b} \omega} \sum_c \epsilon_{abc} B_c \right).
\]  
(A.18)

**Appendix B**

In this appendix, we shall derive the improved Pauli term (3.32) by means of Foldy-Wouthuysen transformation of Dirac equation. The single electron Dirac equation in an ionic crystal and in an external radiation field reads
\[
\frac{i \partial \psi}{\partial t} = H \psi,
\]  
(B.1)

with
\[
H = -i \rho_1 \vec{\tau} \cdot (\vec{\nabla} - ie \vec{A}) + e A_0 + \vec{V}(\vec{r}) \cdot \vec{\tau} + \rho_3 m_e,
\]  
(B.2)

where \((A_0, \vec{A})\) is the four component gauge potential with \(A_0\) containing the self-consistent electrostatic potential of ion cores and electron clouds, \(V(\vec{r})\) is the exchange part of the self-consistent potential, \(\psi(\vec{r})\) is a \(4 \times 1\) spinor wave function and \(\rho_i\) and \(\tau_i\) are the same \(4 \times 4\) matrices introduced in the section 2 (in terms of the standard notation, \(\rho_3 = \beta, \rho_1 \vec{\tau} = \vec{\alpha}\)). Equation (B.1) can be regarded a relativistic Hartree-Fock equation with a filled Dirac sea of negative energy levels and a Fermi sea of the positive levels. As we shall see, to the order we are interested, the details of \(A_0(\vec{r})\) and \(V(\vec{r})\) are unimportant provide that their magnitudes are nonrelativistic and they produce the same conducting band given by (2.28).

For a nonrelativistic electron, the lower \(2 \times 1\) spinor of \(\psi\) is suppressed by a factor \(\sim v/c\) relative to the upper \(2 \times 1\) spinor. Following Foldy and Wouthuysen [12], we decompose the Hamiltonian (B.2) into three part,
\[
H = \rho_3 m_e + \mathcal{E} + \mathcal{O},
\]  
(B.3)
where

\[ E = eA_0(\vec{r}) + \rho_3 \vec{V}(\vec{r}) \cdot \tau \]  \hspace{1cm} (B.4)

\[ O = -i\rho_1 \bar{\tau} \cdot (\vec{\nabla} - ie\vec{A}(\vec{r})) \]. \hspace{1cm} (B.5)

Among sixteen basic $4 \times 4$ matrices, $1, \rho_i, \tau_i, \rho_i \tau_j (i = 1, 2, 3, j = 1, 2, 3),$ $\rho_1, \rho_2, \rho_1 \tau_1$ and $\rho_2 \tau_1$ couple to the upper $2 \times 1$ spinor and the lower one and will be referred to as odd operator, the rest of them does not couple the upper $2 \times 1$ spinor and the lower one and will be referred to as odd operator. Therefore $\rho_3 m_e$ and $E$ is even while $O$ is odd. A systematic nonrelativistic approximation of a Dirac equation amounts to a set of successive unitary transformations

\[ \psi' = e^{-iS} \psi \]  \hspace{1cm} (B.6)

and

\[ H' = H - ie^{-iS} \frac{\partial}{\partial t} e^{-iS} \], \hspace{1cm} (B.7)

which eliminates the odd operators to the desired order of $\frac{1}{m_e}$. For the Dirac Hamiltonian (B.2) and to the order $\frac{1}{m_e}$, the following three transformations serves the purpose:

\[ e^{-iS} = e^{-iS_3} e^{-iS_2} e^{-iS_1} \]  \hspace{1cm} (B.8)

with

\[ S_1 = -i \frac{1}{2m_e} \rho_3 O \]  \hspace{1cm} (B.9)

\[ S_2 = \frac{1}{4m_e^2} (\dot{O} - i[O, E]) \]  \hspace{1cm} (B.10)

and

\[ S_3 = \frac{i}{8m_e^2} \rho_3 \left( \frac{4}{3} \mathcal{O}^3 + \dot{O} + iE, \dot{O} - i[O, E] - i[\dot{O}, E] - i[O, \dot{E}] \right) \]. \hspace{1cm} (B.11)

The transformed Hamiltonian reads

\[ H' = \rho_3 m_e + eA_0 - \frac{1}{2m_e} \rho_3 (\vec{\nabla} - ie\vec{A})^2 - e \rho_3 \bar{\tau} \cdot \vec{B} \]

\[- ie \frac{\vec{\nabla} \cdot \vec{E} + ie}{4m_e^2} \bar{\tau} \cdot \vec{E} \times (\vec{\nabla} - ie\vec{A}) - e \rho_3 \vec{\nabla} \cdot \vec{E} \]

\[ + \frac{e}{8m_e^2} \rho_3 \{ \bar{\tau} \cdot (\vec{\nabla} - ie\vec{A}), \{ \bar{\tau} \cdot (\vec{\nabla} - ie\vec{A}), \vec{\nabla} (\vec{r}) \cdot \bar{\tau} \}\} , \hspace{1cm} (B.12)\]

where $\vec{E} = -\frac{\partial \vec{A}}{\partial t} - \vec{\nabla} A_1$ and $\vec{B} = \vec{\nabla} \times \vec{A}$. If we neglect the local magnetic field produced by the spin ordering, $\vec{A}$ is entirely due to the radiation field, so is $\vec{B}$.
The electric field $\vec{E}$, however, can be decomposed into a static field caused by the ionic potential and the radiation field. The Hamiltonian is a quadratic function of the radiation field, $\vec{A}$, $\vec{E}_{\text{rad}}$, and $\vec{B}$. For a nonrelativistic Fermi sea of plasma frequency $\omega_p$ and a photon of frequency $\omega \sim \omega_p$, $k \sim \omega$. With $|\vec{V}| \sim \omega_p$ and $eA_0(\vec{r}) \sim \omega_p$, it is easy to see that the leading contributions to the antisymmetric part of the self energy function come from the second order perturbation of the Pauli term and the first order perturbation of the term

$$-rac{e^2}{4m_\text{e}} \vec{r} \cdot \vec{A} \times \vec{E}_{\text{rad}}.$$  \hfill (B.13)

of (B.12). The latter gives rise to the Farady angle (3.28).

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