Theory of Orbital Susceptibility on Excitonic Insulator

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We study the temperature dependence of the orbital susceptibility of an excitonic insulator on the basis of a two-band model. It is shown that a drastic change (an anomalous enhancement) in susceptibility as a function of temperature occurs owing to the occurrence of additional orbital susceptibility due to the excitonic gap. We calculate explicitly the temperature dependence of orbital susceptibility for a model of Ta$_2$NiSe$_5$, and show that the result is consistent with experimental results.

In semimetals, electrons and holes form bound pairs of excitons. These bound pairs lead to a nonconducting state called an excitonic insulator. In the 60s much interest was attracted by the excitonic insulator. One of the important and interesting points concerning the excitonic insulator is that the excitonic theory is transformed to the BCS theory by a particle-hole transformation. From the similarity to the BCS theory, the stability of the excitonic insulator, anisotropy, impurity effect, transport properties, and effect of magnetic field have been extensively studied. However, no materials have been identified as an excitonic insulator.

Recently, it has been argued that Ta$_2$NiSe$_5$ is a candidate material of an excitonic insulator from photoemission spectroscopy. In this experiment, the observed energy dispersion seems to be consistent with the theoretical result based on the mean field approximation of a model consisting of electron and hole bands with Coulomb interactions between them. Since this experiment, the stability of the excitonic insulator has been studied on the basis of numerical calculations of the two-band model or an extended model.

Several experiments have been suggested from theoretical viewpoints using the relationship between the excitonic insulator and superconductivity to confirm the existence of the excitonic insulator. For example, the temperature dependence of the ultrasonic absorption coefficient and nuclear magnetic relaxation rate have been calculated. However, there have been no experimental results that confirm the excitonic insulator except for photoemission spectroscopy.

Although this temperature dependence is expected to be a feature of the excitonic insulator, the origin of this drastic change has not been understood theoretically. In this Letter, we study the temperature dependence of orbital susceptibility in a two-band model for the excitonic insulator. We clarify that the temperature dependence of susceptibility is understood as an occurrence of additional orbital susceptibility owing to the excitonic phase transition. We show that the obtained temperature dependence of orbital susceptibility agrees very well with experiments.

To study the orbital susceptibility of the excitonic insulator, we use a two-band model consisting of electron and hole bands with Coulomb interactions under a magnetic field. We use a recently developed exact formalism for orbital susceptibility. A schematic picture of this model is shown in Fig. 1, where $\alpha$ is the index of two chains $\alpha = 1, 2$, and $\alpha$ is the lattice constant between two sites. Red and blue circles indicate the lattice sites, and $V$ indicates the Coulomb interaction between chains.

First, we construct a Hamiltonian of the effective model of Fig. 1. A one-body Hamiltonian under the magnetic field is given by

$$\hat{H}(\mathbf{r}) = \frac{1}{2m}(\mathbf{p} - e\mathbf{A}(\mathbf{r}))^2 + \sum_{\alpha,j} V(\mathbf{r}_{\alpha,j}),$$

where $m$ and $e$ ($e < 0$) are the mass and charge of an electron, respectively, $c$ is the speed of light, $\mathbf{A}(\mathbf{r})$ is a vector potential, and $V(\mathbf{r}_{\alpha,j})$ is a potential at $\mathbf{r}_{\alpha,j} = \mathbf{r} - \mathbf{R}_{\alpha,j}$, where $\mathbf{R}_{\alpha,j}$ is the position of the $j$-th site of the $\alpha$ chain.

The wave function under the magnetic field is obtained as

$$\Phi(\mathbf{r}_{\alpha,j}) = \exp \left( \frac{ie}{\hbar} \mathbf{A}(\mathbf{r}_{\alpha,j}) \cdot \mathbf{r} \right) \phi(\mathbf{r}_{\alpha,j}),$$

where $\mathbf{A}(\mathbf{r}) = \mathbf{A}(\mathbf{R}_{\alpha,j})$. $\phi(\mathbf{r}_{\alpha,j})$ is an eigenstate of

$$\left[ \frac{1}{2m} \mathbf{p}^2 + V(\mathbf{r}_{\alpha,j}) \right] \phi(\mathbf{r}_{\alpha,j}) = \epsilon_0 \phi(\mathbf{r}_{\alpha,j}),$$

where $\epsilon_0$ is an eigenvalue. We assume the orthogonality of the wave functions

$$\int d\mathbf{r} \Phi^*(\mathbf{r}_{\alpha,j}) \Phi(\mathbf{r}'_{\alpha',j}) = \delta_{\alpha\alpha'} \delta_{jj'},$$

for simplicity.

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Fig. 1. (Color online) Schematic picture of two-chain model. A magnetic field is applied to the $z$-axis. Red and blue circles indicate the lattice sites, and $V$ is the Coulomb interaction between two chains.
The term of chains, which is expressed as gauges of Eq. (9) into Eq. (7), the Peierls phases are obtained

\[ H = \frac{1}{2m}(p - e\mathbf{A}(\mathbf{r}) - A_{\alpha',j})^2 + \sum_{\alpha,j} V(\mathbf{r},\alpha,j). \]

where \( A(\mathbf{r}) = \left\{ \begin{array}{ll}
(Hz, 0, 0) & \text{for } y \text{ direction}, \\
(Hx, 0) & \text{for } z \text{ direction},
\end{array} \right. \)

\[ \Phi_{ij}^{\alpha\alpha'} = \frac{e}{c} (A_{\alpha,i} - A_{\alpha',j}) \cdot \frac{\mathbf{R}_{\alpha,i} + \mathbf{R}_{\alpha',j}}{2}, \quad (9) \]

Here, \( \Phi_{ij}^{\alpha\alpha'} \) and \( \chi_{ij}^{\alpha\alpha'} \) are the Peierls phase and additional phase, respectively. \( \chi_{ij}^{\alpha\alpha'} \) is discussed in Ref. 20. In order to study the magnetic field along the y- and z-axes, we use Landau gauges as follows.

\[ \chi_{ij}^{\alpha\alpha'} = \begin{cases} 0 & \text{for } y \text{ direction}, \\
\frac{\partial}{\partial z}(x_{\alpha,i} - x_{\alpha',j})(y_{\alpha,i} + y_{\alpha',j}) & \text{for } z \text{ direction}, \end{cases} \quad (10) \]

where \( x_{\alpha,i} \) and \( y_{\alpha,i} \) are the x- and y-components of \( \mathbf{r} - \mathbf{R}_{\alpha,i} \), respectively. \( \chi_{ij}^{\alpha\alpha'} \) is obtained in the same way.

Since the general formula of the matrix elements of Eq. (5) is very complicated, \( \Phi_{ij}^{\alpha\alpha'} = (-1)^\alpha \delta_{\alpha,\alpha'}(t_0 + \frac{e}{c} t_0 h^2), \) (11)

where \( t_0 \) and \( t_0 \) are the transfer integral and the correction of the transfer integral under the magnetic field, respectively. The detail of \( t_2 \) is discussed in Ref. 20. \( h \) is the dimensionless magnetic field defined as

\[ h = \frac{eH}{2c\hbar}. \]

For \( i = j = \alpha = \alpha' \), the matrix element is

\[ \text{Eq. (5)} \approx (-1)^\alpha \delta_{\alpha,\alpha'}(t_0 + \frac{e}{c} t_0 h^2), \]

where \( e \) and \( \mu \) are a one-body level and a chemical potential, respectively, and \( e_2 \) is the coefficient of \( \hbar^2 \), which comes from the term of \( \mathbf{A}(\mathbf{r}) - \mathbf{A}_{\alpha,j} \).

Next, we study the Coulomb interaction \( V \) between two chains, which is expressed as

\[ V = \int d\mathbf{r}_1 d\mathbf{r}_2 \Phi^*(\mathbf{r}_1) \Phi^*(\mathbf{r}_2) \frac{e^2}{|\mathbf{R}_1 - \mathbf{R}_2|} \Phi(\mathbf{r}_1) \Phi(\mathbf{r}_2). \]

By substituting Eq. (2), it is found that \( V \) is independent of the magnetic field.

As a result, the Hamiltonian of an effective model of Fig. 1 becomes

\[ H = \sum_i \left[ (-\mu - e_2 \hbar^2)n_{i,j} + (\mu - e_2 \hbar^2)n_{i,j} \right] + V \sum_{n_{i,j} = 0} \left[ n_{i,j} + 2 \right]. \]

\[ H = \frac{1}{2m}(p - e\mathbf{A}(\mathbf{r}) - A_{\alpha',j})^2 + \sum_{\alpha,j} V(\mathbf{r},\alpha,j). \]

\[ \Phi_{ij}^{\alpha\alpha'} = \frac{e}{c} (A_{\alpha,i} - A_{\alpha',j}) \cdot \frac{\mathbf{R}_{\alpha,i} + \mathbf{R}_{\alpha',j}}{2}, \quad (7) \]

\[ \chi_{ij}^{\alpha\alpha'} = -\frac{e}{c} (A_{\alpha,i} - A_{\alpha',j}) \cdot (\mathbf{r} - \mathbf{R}_{\alpha,i} + \mathbf{R}_{\alpha',j}). \quad (8) \]

By substituting the Coulomb interaction \( V \) between two chains, which is expressed as

\[ V = \int d\mathbf{r}_1 d\mathbf{r}_2 \Phi^*(\mathbf{r}_1) \Phi^*(\mathbf{r}_2) \frac{e^2}{|\mathbf{R}_1 - \mathbf{R}_2|} \Phi(\mathbf{r}_1) \Phi(\mathbf{r}_2). \]

By substituting Eq. (2), it is found that \( V \) is independent of the magnetic field.
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In a similar way, under the magnetic field along the z-axis, the

Fig. 3. (Color online) e dependence on the transition temperature at V/t_0 = 2.0.

becomes maximum near the band edge.

Next, we calculate the orbital susceptibility in this system. As discussed in Eq. (11), the transfer integral is modified by the magnetic field along the y-axis as \( t(h) = t_0 + t_2 h^2 \), while the transfer integral modified by that along the z-axis includes the Peierls phase. The effective Hamiltonian under the magnetic field along the y-axis is given by replacing the \( \zeta_i \) of Eq. (19) with \( \zeta_{ik} = 2(-t_0 + t_2 h^2) \cos k_x, a - e \). As a result, the eigenvalues are obtained as

\[
E_{k,\pm} = -\mu + e_2 h^2 \pm \sqrt{\zeta_{ik}^2 + \Delta_{EX}}.
\]

In a similar way, under the magnetic field along the z-axis, the effective Hamiltonian is given by

\[
H_{tot} = \sum_k \left[ (-\zeta_{ik} - \mu + e_2 h^2)c_{i,k}^\dagger c_{i,k}^\dagger + \zeta_{ik} c_{i,k}^\dagger c_{i,k}^\dagger + \Delta_{EX} c_{i,k}^\dagger c_{i,k} + \Delta_{EX} c_{i,k}^\dagger c_{i,k}^\dagger \right] + \frac{N\Delta_{EX}^2}{V},
\]

where \( \zeta_{ik} \) and \( \zeta_{ik}^\dagger \) are obtained as

\[
\zeta_{ik} = 2(-t_0 + t_2 h^2) \cos (k_x + 2h) + e.
\]

The eigenvalues of Eq. (24) are given by

\[
E_{k,\pm} = -\mu + e_2 h^2 + \sqrt{\left(\zeta_{ik} - \zeta_{ik}^\dagger\right)^2 + 4\Delta_{EX}^2}. \tag{27}
\]

The thermodynamic potential of the system under the magnetic field, \( \Omega_{tot}(H) \), is defined as

\[
e^{-\beta V_{tot}(H)} = \text{Tr} \exp(-\beta H_{tot}). \tag{28}
\]

By substituting the Hamiltonian, the thermodynamic potential is obtained as

\[
\Omega_{tot}(h) = \frac{N\Delta_{EX}^2}{V} - \frac{1}{\beta} \sum_{k,\pm} \ln (1 + e^{-\beta E_{k,\pm}}). \tag{29}
\]

The orbital susceptibility is calculated from

\[
\chi = -\frac{1}{H} \left[ \frac{\partial \Omega_{tot}(H)}{\partial H} \right]_{H=0} = -\frac{e^2 d^4}{4c^2 h^2} \left[ \frac{1}{h} \frac{\partial \Omega_{tot}(h)}{\partial h} \right]_{h=0}. \tag{30}
\]

where the chemical potential is always fixed at \( \mu = 0 \) because we discuss only the half-filling.

Figure 4 shows the temperature dependence on the orbital susceptibility at V/t_0 = 2.0, t_2/t_0 = -0.2, \( \epsilon/t_0 = 0.1 \), and \( \epsilon/t_0 = 3.0, 2.0, 1.0, \) and 0.0. The magnetic field is applied along the y-axis.

\[
\zeta_{2,k} = 2(-t_0 + t_2 h^2) \cos (k_x + 2h) + e. \tag{26}
\]

The eigenvalues of Eq. (24) are given by

\[
E_{k,\pm} = -\mu + e_2 h^2 + \sqrt{\left(\zeta_{ik} - \zeta_{ik}^\dagger\right)^2 + 4\Delta_{EX}^2}. \tag{27}
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\[
\zeta_{2,k} = 2(-t_0 + t_2 h^2) \cos (k_x + 2h) + e. \tag{26}
\]
ceptibility increases negatively.

In order to understand the temperature dependence on the susceptibility in detail, we divide the susceptibility into three components $\chi_{\text{PP}}, \chi_{\text{t}},$ and $\chi_{\text{core}}$, where $\chi_{\text{PP}}, \chi_{\text{t}},$ and $\chi_{\text{core}}$ are the susceptibilities coming from the Peierls phase, $t_2$, and $e_2$, respectively. The total susceptibility is obtained as

$$\chi = \chi_{\text{PP}} + \chi_{\text{t}} + \chi_{\text{core}}.$$  \hfill (31)

These susceptibilities are calculated as follows.\(^{20}\) For $\chi_{\text{PP}}$, the magnetic field is applied along the $z$-axis, and the parameters are set as $V/t_0 = 2.0, t_2/t_0 = 0.0, \text{and } e_2/t_0 = 0.0$. In the same way, for $\chi_{\text{t}}$ ($\chi_{\text{core}}$), the magnetic field is applied along the $y$-axis, and the parameters are set as $V/t_0 = 2.0, t_2/t_0 = -0.2, \text{and } e_2/t_0 = 0.0$ ($V/t_0 = 2.0, t_2/t_0 = 0.0, \text{and } e_2/t_0 = 0.1$).

Figure 6 shows the temperature dependence on the susceptibilities of $\chi_{\text{PP}}, \chi_{\text{t}}, \chi_{\text{core}},$ and $\chi$ at $\epsilon/t_0 = 2.0$. It is found that above the transition temperature, the temperature dependence on the total susceptibility comes from $\chi_{\text{t}}$. It is also found that below the transition temperature, the temperature dependence is derived from $\chi_{\text{PP}}$. Although the temperature dependence of $\chi_{\text{t}}$, also changes below the transition temperature, $\chi_{\text{t}}$ is independent of temperature in this case. We discuss the physical meaning of the temperature dependences of $\chi_{\text{core}}, \chi_{\text{t}},$ and $\chi_{\text{PP}}$. Since $\chi_{\text{core}}$ is the atomic diamagnetism, it is independent of the temperature, as shown in Fig. 6. The origin of $\chi_{\text{t}}$ is the correction of the transfer integral under the magnetic field as discussed in Eq. (11). Above the transition temperature, the valence band and conduction band touch at $k_a = \pi$ and $-\pi$. Since the energy gain owing to the correction of the transfer integral is zero at the zero temperature of the normal state, the origin of the temperature dependence of $\chi_{\text{t}}$ is a thermal excitation. When the excitonic gap occurs below the transition temperature, the susceptibility due to the thermal excitation becomes smaller. On the other hand, the mixing between the conduction band and the valence band occurs below the transition temperature. As a result, it is expected that a finite energy gain owing to the correction of the transfer integral occurs and it is the origin of $\chi_{\text{t}}$. However, the details of the temperature dependence are still unclear and this is a future research topic. The origin of the finite contribution of $\chi_{\text{PP}}$ below the transition temperature is due to the coherent hopping between chains induced by the formation of the excitonic state.

As a result, the effect of the Peierls phase flowing between two chains increases as the temperature decreases.

Finally, we compare the present theoretical result with experiments on Ta$_2$NiSe$_5$. As discussed in Introduction, the temperature dependence of the susceptibility of Ta$_2$NiSe$_5$ has been reported.\(^{17}\) As the temperature decreases from 900 K, the susceptibility decreases almost linearly, and the susceptibility decreases drastically below 400 K. The susceptibility becomes independent of temperature below 200 K. When we set the hopping parameter as $t_0 \sim 0.2$ eV, the present result $\chi$ of Fig. 6 is consistent with the temperature dependence of Ta$_2$NiSe$_5$. Recently, the electronic state of Ta$_2$NiSe$_5$ has been studied in detail, and the effective model is known to be a three-band model.\(^{15,16}\) Although the essential point of the orbital susceptibility is understood on the basis of the two-band model in this paper, it is necessary to discuss the orbital susceptibility on the basis of the three-band model in order to understand the orbital susceptibility of Ta$_2$NiSe$_5$ in detail.

In conclusion, we studied the temperature dependence of orbital susceptibility on the excitonic insulator on the basis of a two-band model with the magnetic field. We clarified that the temperature dependence of the susceptibility is derived from the occurrence of additional orbital susceptibility due to the excitonic gap. We calculated the temperature dependence of orbital susceptibility and the result is consistent with the experimental result of Ta$_2$NiSe$_5$.

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