Anomalous Spin Response in Non-centrosymmetric Compounds

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We examine static spin susceptibilities $\chi_{\alpha\beta}(\mathbf{q})$ of spin components $S_\alpha$ and $S_\beta$ in the non-centrosymmetric tetragonal system. These show anomalous momentum dependences like $\chi_{xx}(\mathbf{q}) - \chi_{yy}(\mathbf{q}) \sim q_x^2 - q_y^2$ and $\chi_{xy}(\mathbf{q}) + \chi_{yx}(\mathbf{q}) \sim q_x q_y$, which vanish in centrosymmetric systems. The magnitudes of the anomalous spin susceptibilities are enhanced by the on-site Coulomb interaction, especially, around an ordering wave vector. The significant and anomalous momentum dependences of these susceptibilities are explained by a group theoretical analysis. As the direct probe of the anomalous spin susceptibility, we propose a polarized neutron scattering experiment.

Recently, the non-centrosymmetric heavy fermion superconductor attracts much attention. For a tetragonal compound CePt$_3$Si, which has been intensively studied from both experimental and theoretical sides, the superconducting transition has been observed at 0.75K below the antiferromagnetic transition temperature 2.2K at ambient pressure [1]. In addition, CeRhSi$_3$ and CeSi$_3$ also show superconductivity by applying pressure [2,3]. Commonly, superconductivity in these compounds has been found around the antiferromagnetic phase like in centrosymmetric heavy electron systems [1,2,3,4].

Theoretically, the non-centrosymmetric system is characterised by the Rashba-type effective spin-orbit coupling, which has an antisymmetric momentum dependence with respect to the spatial inversion [5,6]. As a characteristic feature of the model, it has been shown that in terms of a band splitting caused by the spin-orbit coupling, the uniform susceptibility has large Van-Vleck type contribution [2]. Furthermore, the thermal average of the spin operator of an electron with a momentum $\mathbf{k}$ does not necessarily vanish [5,6], although any magnetic moment disappearance by canceling out spins in the paramagnetic state. The effect of electric spins on transport coefficients like spin Hall effect are intensively studied in the field of spintronics [7,8,9].

For the normal state property of non-centrosymmetric heavy fermion superconductors, the difference from centrosymmetric systems by the Rashba-type spin-orbit coupling has not been suggested except for the quantities mentioned above. Considering that in hydrostatic pressure, the superconductivity appears from the paramagnetic state with decreasing temperature, the spin fluctuation will be one of keys of superconductivity, and a normal state property characterising the non-centrosymmetric compound may relate with the mechanism of the superconductivity. Furthermore, as well as transport coefficients, it is expected that the remaining electron spins affect the magnetic excitation in the non-centrosymmetric compound.

In this Letter, we examine static spin susceptibilities $\chi_{\alpha\beta}(\mathbf{q})$ of $\alpha$- and $\beta$-components of spin operators, based on a simple Hubbard model including the Rashba-term. Significant momentum dependences like $\chi_{xx}(\mathbf{q}) - \chi_{yy}(\mathbf{q}) \sim q_x^2 - q_y^2$ and $\chi_{xy}(\mathbf{q}) + \chi_{yx}(\mathbf{q}) \sim q_x q_y$ will be shown, where these susceptibilities vanish in the centrosymmetric system. In order to clarify the origin of momentum dependences, we carry out a group theoretical analysis to show that the symmetry of momentum dependence of $\chi_{\alpha\beta}(\mathbf{q})$ is identical with the representation of the product of spin operators included in a corresponding susceptibility. In order to observe the unusual momentum dependence of spin susceptibilities, we suggest a polarized neutron scattering experiment, especially, around the magnetic instability.

In the following, we describe a non-centrosymmetric system by the following Hamiltonian

$$ H = H_0 + H_1, $$

$$ H_0 = \sum_{k\sigma\sigma'} [ (\varepsilon_k - \mu) \sigma_0 + \mathbf{g}_k \cdot \hat{n} ]_{\sigma\sigma'} c^\dagger_{k\sigma} c_{k\sigma'}, $$

$$ H_1 = U \sum_i n_{i\uparrow} n_{i\downarrow}, $$

where $\varepsilon_{k\sigma}$ and $c^\dagger_{k\sigma}$ are annihilation and creation operators of an electron with a momentum $\mathbf{k}$ and a spin $\sigma$. Here, $\varepsilon_{\mathbf{k}}$ and $\mu$ are the energy dispersion of electrons and the chemical potential, respectively, while $\mathbf{g}_k$ describes the Rashba field satisfying $\mathbf{g}_k = -g_{\mathbf{k}} \cdot \mathbf{k}$, which breaks the inversion symmetry. Then, eigenenergies of the non-interacting part are given by $\varepsilon_{k\pm} = \varepsilon_k \pm |\mathbf{g}_k| - \mu$. In addition, $H_1$ is the on-site interaction term.

In the non-interacting case, the electron Green’s function is given by

$$ \hat{G}_0^{(0)}(\mathbf{k}, \omega_n) = G_+^{(0)}(\mathbf{k}, \omega_n) \sigma_0 + \frac{\mathbf{g}_k}{|\mathbf{g}_k|} \cdot \hat{n} \hat{G}_-^{(0)}(\mathbf{k}, \omega_n), $$

with $G_\pm^{(0)}(\mathbf{k}, \omega_n) = \frac{1}{2} \{ (\omega_n - \varepsilon_{k\pm})^{-1} \pm (\omega_n - \varepsilon_{k-\pm})^{-1} \}$, where $\omega_n$ is a fermionic Matsubara frequency. Here, since $G_-^{(0)}(\mathbf{k}, \omega_n)$ is expanded only in odd-power of $|\mathbf{g}_k|$, $G_-^{(0)}(\mathbf{k}, \omega_n)$ vanishes in the limit of $|\mathbf{g}_k| \rightarrow 0$. Summing up the Green’s function with respect to the Matsubara frequency, the first term gives electron densities of $(\mathbf{k}, \pm)$-states, which determine shapes of two Fermi surfaces, while the second term contributes electron spins of $(\mathbf{k}, \pm)$-states, whose directions are opposite from each
FIG. 1: Schematic views of charges (a) and spins (b) of electrons. Red and blue lines denote Fermi surfaces of $\varepsilon_k^-$ and $\varepsilon_k^+$, respectively. Red and blue wedges are spins of electrons belonging to $\varepsilon_k^-$ and $\varepsilon_k^+$, respectively. Spins of $\varepsilon_k^-$ electrons remain in the region between two Fermi surfaces.

other due to the minus sign in front of $(i\omega_n - \varepsilon_{k^-})^{-1}$ of $G^{(0)}(k, \omega_n)$. These features of electrons are schematically drawn in Figs. 1(a) and 1(b), respectively. From Fig. 1(b), it is understood that spins of electrons belong to $\varepsilon_k^-$ band remain in the region between two Fermi surfaces, while a spin of an $\varepsilon_k^+$-band electron at a $k$ cancels out another spin of an $\varepsilon_k^+$-band electron at the same $k$ within the inner Fermi surface. It is expected that the remaining electric spin affects the magnetic response.

In order to examine the magnetic excitation in the non-centrosymmetric system, we consider the dynamical susceptibility, defined as

$$\chi_{\alpha\beta}(q, i\Omega_n) = \int_0^\beta d\tau e^{i\Omega_n\tau} \langle T_\tau[(S^\alpha_q(\tau) - \langle S^\alpha_q \rangle)(S^\beta_q(0) - \langle S^\beta_q \rangle)] \rangle \tag{5}$$

where $\langle \cdots \rangle$ means the thermal average of $\cdots$, $T_\tau$ denotes the imaginary-time chronological ordering operator, and $\Omega_n$ is a bosonic Matsubara frequency. Here, charge and spin operators with a wave vector $q$ are given by $S^\alpha_q = 1/2 \sum_{k\sigma} c\gamma_k c_{k+q\sigma}$ and $S^\alpha_q = \sum_{k\sigma} \sigma^\alpha_{\sigma\gamma} c\gamma_k c_{k+q\sigma}$ ($\alpha = x, y, z$), respectively, where $\delta^\alpha$ is an $\alpha$-component of Pauli matrices. In centrosymmetric systems, all off-diagonal susceptibilities disappear. On the other hand, it should be noted that off-diagonal susceptibilities do not always vanish in non-centrosymmetric systems[1], and even susceptibilities between spin and charge operators $\chi_{\alpha\beta}(q, i\Omega_n)$ and $\chi_{\alpha\alpha}(q, i\Omega_n)$ ($\alpha = x, y, z$) have non-zero values for $\Omega_n \neq 0$.

According to the diagrammatic technique, we formulate $\chi_{\alpha\beta}(q, i\Omega_n)$ with use of the Green’s function $G^{(0)}(k, \omega_n)$ given above. In the non-interacting case, the susceptibility $\chi_{\alpha\beta}^{(0)}(q, i\Omega_n)$ is calculated through a transformation from $\tilde{\chi}_{\alpha\beta\sigma\sigma\sigma\sigma'}(q, i\Omega_n)$ defined by,

$$\tilde{\chi}_{\alpha\beta\sigma\sigma\sigma\sigma'}(q, i\Omega_n) = -T \sum_k \sum_m G_{\sigma\sigma'}^{(0)}(k, \omega_m) G_{\sigma\sigma'}^{(0)}(q + k, \omega_m + i\Omega_n) \tag{6}$$

which corresponds to the one-bubble diagram [10]. In the interacting case with a finite $U$, the expression of

FIG. 2: (a) $q$-dependences of static susceptibilities, $\chi_{xx}^{(0)}(q, 0)$, $\chi_{xy}^{(0)}(q, 0)$, $\chi_{yx}^{(0)}(q, 0)$, and $(\chi_{xx}^{(0)}(q, 0) - \chi_{yy}^{(0)}(q, 0))/2$, where insets show Fermi surfaces in the first quadrant of the Brillouin zone (left) and the path in $q$-space (right). (b) $q$-dependences of static susceptibilities, $\chi_{xx}^{(0)}(q, 0)$, $\chi_{xy}^{(0)}(q, 0)$, and $\chi_{yz}^{(0)}(q, 0)$ with the momentum path shown in the inset.

$\chi_{\alpha\beta}(q, i\Omega_n)$ is given within the random phase approximation (RPA) [11, 12, 13] as

$$\hat{\chi}(q, i\Omega_n) = \left[1 - 2\hat{\chi}^{(0)}(q, i\Omega_n)\hat{U}\right]^{-1} \hat{\chi}^{(0)}(q, i\Omega_n), \tag{7}$$

where matrix elements of $\hat{U}$ are given by $U_{\alpha\beta} = \delta_{\alpha\beta} U_{\alpha\alpha}$ with $U_{xx} = -U$ and $U_{zz} = U_{yy} = U$.

Then, we calculate $\chi_{\alpha\beta}(q, i\Omega_n)$ numerically, based on a two-dimensional non-centrosymmetric system with an energy dispersion $\varepsilon_k = 2t_1(\cos k_x + \cos k_y) + 4t_2 \cos k_x \cos k_y$ and a Rashba-field $gk = g(\sin k_y, -\sin k_x, 0)$. For parameters, $t_2/t_1=0.35$ and $g/t_1=0.2$ are chosen, we can reproduce quasi two dimensional Fermi surfaces of CePt$_3$Si obtained by the band calculation[14, 15, 16]. In Fig. 2, we show momentum dependences of static spin susceptibilities in the non-interacting case. Diagonal components of $\tilde{\chi}^{(0)}(q)$ are shown in Fig. 2(a), where Fermi surfaces and the momentum path are also shown in insets. Unlike in centrosymmetric systems, all momentum dependences of $\chi_{xx}^{(0)}(q)$, $\chi_{xy}^{(0)}(q)$, $\chi_{yz}^{(0)}(q)$, and $\chi_{yy}^{(0)}(q)$ are different from each other. Especially, in a path from $(0, \pi)$ to $(\pi, 0)$, $\chi_{xx}^{(0)}(q) - \chi_{yy}^{(0)}(q)$ is antisymmetric around the midpoint $(\pi/2, \pi/2)$, and it vanishes in a diagonal path from $(\pi, \pi)$ to $(0, 0)$. Therefore, the momentum dependence
of \( \chi^{(0)}_{xx}(q) - \chi^{(0)}_{yy}(q) \) has the typical \( q_x^2 - q_y^2 \) symmetry. Likewise, momentum dependences of \( \chi^{(0)}_{yy}(q) + \chi^{(0)}_{xx}(q) \), \( (\chi^{(0)}_{yy}(q) - \chi^{(0)}_{yy}(q))/i \), and \( (\chi^{(0)}_{xx}(q) - \chi^{(0)}_{xx}(q))/i \) are shown in Fig. 2(b), where the momentum path is depicted in the inset. In the figure, \( \chi^{(0)}_{yy}(q) + \chi^{(0)}_{xx}(q) \) is symmetric around \((0,0)\) and \((2\pi,0)\), while it is antisymmetric around \((\pi,\pi)\). Accordingly, the momentum dependence of \( \chi^{(0)}_{xy}(q) + \chi^{(0)}_{yx}(q) \) is a \( q_xq_y \)-type. Similarly, it can be understood easily that momentum dependences of \( (\chi^{(0)}_{yy}(q) - \chi^{(0)}_{yy}(q))/i \) and \( (\chi^{(0)}_{xx}(q) - \chi^{(0)}(q))/i \) are \( q_y \)- and \( q_x \)-types, respectively. Thus, spin susceptibilities have significant and unusual momentum dependences related with corresponding spin indices.

For an intuitive picture of anomalous momentum dependencies of spin susceptibilities, we consider particle-hole excitations in the non-centrosymmetric system. According to the prescription to calculate \( \chi_{\alpha\beta}(q) \), contributions to \( \chi^{(0)}_{yy}(q) \) and \( \chi^{(0)}_{xx}(q) \) are separated to two terms of convolutions, \( T \sum_{s,n} G^+(k, \omega_m) G^+(-k + q, \omega_m + i\alpha_n) \) and \( T \sum_{s,n} G^0(k, \omega_m) G^0(-k + q, \omega_m + i\alpha_n) \), where the second part vanishes in the limit of the zero Rashba field. Here, we show schematic figures of these particle-hole excitations in Figs. 3(a) and 3(b), respectively. In Fig. 3(a), the mechanism of particle-hole excitations is not different from that of the centrosymmetric case. In this case, the contribution to \( \chi^{(0)}_{xx}(q) \) is isotropic in the spin space. On the other hand, a particle-hole excitation with a momentum transfer \( q_x \) shown in Fig. 3(b) only contributes to \( \chi^{(0)}_{yy}(q_x) \) for low energy excitations: rotating the quantization axis to the \(-e_y \) direction, the longitudinal spin excitation is only permitted, and other excitations are almost forbidden because of the energy loss from spin rotations against the Rashba-field. Similarly, if we consider a particle-hole excitation with a momentum transfer \( q_y \) shown in Fig. 3(b), only \( \chi^{(0)}_{xx}(q_y) \) has the contribution. This is consistent with a behavior in a path \((0,0)-(0,\pi)\) of Fig. 2(a), where the momentum dependence of \( \chi^{(0)}_{xx}(q) \) is considerably different from those of \( \chi^{(0)}_{yy}(q) \) and \( \chi^{(0)}_{xx}(q) \).

Considering these points, \( \chi^{(0)}_{xx}(q) - \chi^{(0)}_{yy}(q) \) should have the \( q_x^2 - q_y^2 \)-type momentum dependence. Similar consideration will also work well for \( \chi^{(0)}_{xy}(q) + \chi^{(0)}_{yx}(q) \).

The above discussion is for the non-interacting case. By the interaction \( U \), magnitudes of anomalous \( \chi_{xx}(q) - \chi_{yy}(q) \) and \( \chi_{xy}(q) + \chi_{yx}(q) \) should be changed. In order to investigate the effect of interaction on \( q \)-dependences of susceptibilities, we calculate \( \chi_{\alpha\beta}(q) \) within RPA using the same parameter set as in Fig. 2. With the critical interaction \( U_c = 2.551t_1 \), the paramagnetic state becomes unstable at an ordering wave vector \( Q = (0.81\pi, 0.25\pi) \).

We show momentum dependences of \( \chi_{zz}(q) \), \( \chi_{xx}(q) + \chi_{yy}(q)/2 \), and anomalous \( \chi_{xx}(q) - \chi_{yy}(q)/2 \) in Figs. 4(a), 4(b), and 4(c), respectively. \( \chi_{zz}(q) \) and \( \chi_{xx}(q) + \chi_{yy}(q)/2 \) are enhanced with increasing \( U \), but have different \( q \)-dependences from each other. Furthermore, the amplitude of \( \chi_{xx}(q) - \chi_{yy}(q)/2 \) increases considerably around the ordering wave vector \( Q \) in comparison with the corresponding value of the non-interacting case. In addition, we note that the amplitude of \( \chi_{xy}(q) + \chi_{yx}(q) \)
TABLE I: Classification of spin product $S^{a}S^{b}$ according to irreducible representations of the $C_{4v} \times K$ group. Then, $S^{a}$, $S^{b}$, and $\{S^{a}, S^{b}\}$ belong to $A_{1}^{+}$, $A_{2}^{-}$, and $E^{-}$ irreducible representations, respectively. The third column shows the basis function in the momentum space. The superscript + (-) of irreducible representations expresses the even (odd) parity with respect to the time reversal.

| $\Gamma$ | $S^{a}S^{b}$ | basis |
|----------|--------------|-------|
| $A_{1}^{+}$ | $S^{a}S^{b}$ | $q_{x}^{2}$, $q_{x}^{2} + q_{y}^{2}$ |
| $A_{2}^{-}$ | $i(S^{a}S^{b} - S^{b}S^{a})$ | $q_{x}q_{y}(q_{x}^{2} - q_{y}^{2})$ |
| $B_{1}^{+}$ | $S^{a}S^{b} - S^{b}S^{a}$ | $q_{x}^{2} - q_{y}^{2}$ |
| $B_{2}^{+}$ | $S^{a}S^{a} + S^{b}S^{b}$ | $q_{x}q_{y}$ |
| $E^{+}$ | $\{i(S^{a}S^{b} - S^{b}S^{a}), i(S^{a}S^{b} - S^{b}S^{b})\}$ | $\{q_{x}q_{y}, q_{x}q_{y}\}$ |
| $A_{2}^{-}$ | $S^{a}S^{a} + S^{b}S^{b}$ | $q_{x}q_{y}(q_{x}^{2} - q_{y}^{2})$ |
| $E^{-}$ | $\{i(S^{a}S^{a} - S^{b}S^{b}), i(S^{a}S^{b} - S^{b}S^{b})\}$ | $\{q_{x}q_{y}\}$ |

is also enhanced with increasing $U$.

In order to clarify the origin of the unusual momentum dependence of the spin susceptibility, we carry out a group theoretical analysis. In the present case, the relevant group is $C_{4v} \times K$, where $C_{4v}$ is the tetragonal point group without the inversion symmetry, and $K$ is the time-reversal symmetry group. From the definition, the momentum dependence of $\chi_{\alpha\beta}(q, i\Omega_{n})$ is determined by the expectation value of the right hand side. Noting that both spin and momentum are transformed by symmetric operations of the group, the representation of the operator in the expectation value is $\Gamma_{s} \otimes \Gamma_{m}$, where $\Gamma_{s}$ and $\Gamma_{m}$ are representations of the spin product and the momentum dependence, respectively. Since the operator giving a non-zero expectation value $\langle \hat{O} \rangle$ belongs to the identity representation $\Gamma_{1}$ ($A_{1}^{+}$ in Table I) in the group, the relation $\Gamma_{s} \otimes \Gamma_{m} = \Gamma_{1}$ is obtained, so that finally

$$
\Gamma_{m} = \Gamma_{s}
$$

(8)
is required. For the symmetry of the $q$-dependence of $\chi_{\alpha\beta}(q, i\Omega_{n})$, it is sufficient to know the representation of the spin-product $S^{a}S^{b}$, where the classification of the product is given in Table I. Therefore, representations of $q$-dependences of $\chi_{xx}(q) - \chi_{yy}(q)$ and $\chi_{xy}(q) + \chi_{yx}(q)$ are $B_{1}^{+}$ and $B_{2}^{+}$, respectively. These results are consistent with $q$-dependences of anomalous susceptibilities shown in Figs. 2 and 4. Thus, it is shown that the symmetry of the $q$-dependence of $\chi_{\alpha\beta}(q, i\Omega_{n})$ is identical with the representation of the spin-product $S^{a}S^{b}$.

Finally, we suggest a polarized neutron scattering experiment to observe the novel anomalous momentum dependence of susceptibility in non-centrosymmetric heavy fermion systems, where even anomalous susceptibilities will be enhanced by the strong interaction as shown in Fig. 4. Among non-centrosymmetric heavy fermion compounds, the only available neutron scattering data are for magnetic structures of CePt$_{3}$Si$_{3}$[17] and CeRhSi$_{3}$[18]. For CePt$_{3}$Si, considering the ordering wave vector $(0,0,\pi)$, to observe the anomalous momentum dependence of susceptibilities will be difficult, because the symmetry of $q$-dependence of every anomalous susceptibility is not the $A_{1}^{+}$ representation. On the other hand, for CeRhSi$_{3}$, whose ordering wave vector is $(\pm 0.43\pi,0,\pi)$, it will be more promising to observe the anomalous momentum dependence of, especially, $\chi_{xx}(q) - \chi_{yy}(q)$ just above the magnetic transition temperature, since the susceptibility will be enhanced around the ordering wave vector by approaching the magnetic transition temperature.

In summary, we have examined the dynamical susceptibility in the non-centrosymmetric tetragonal system. It has been shown that affected by the Rashba term, $\chi_{xx}(q) - \chi_{yy}(q)$ and $\chi_{xy}(q) + \chi_{yx}(q)$ have $q_{x}^{2} - q_{y}^{2}$ and $q_{x}q_{y}$-types of momentum dependences, respectively. The group theoretical analysis has been used to explain the peculiar feature in the non-centrosymmetric systems. Since the unusual susceptibility is also enhanced by the Coulomb interaction, it is desirable to observe the unusual momentum dependence of susceptibility by the polarized neutron scattering experiment.

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