Mechanism of Resonant X-Ray Scattering in DyB_2C_2

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Abstract

For elucidating the relation between the resonant x-ray scattering (RXS) signal and the orbital order, we calculate the RXS spectra near the Dy $L_{\rm III}$ absorption edge in DyB_2C_2, in the antiferro-quadrupole (AFQ) ordering phase. Using a microscopic model that the $4f$ states of Dy are atomic while the $5d$ states form an energy band with a reasonable density of states, we obtain sufficient RXS intensities on the AFQ superlattice spots assuming the quadrupole order in the $4f$ states but without any lattice distortion, in good agreement with the recent experiment. The present result demonstrates the mechanism that the intensity is brought about by the modulation of $5d$ states through the anisotropic term of the $5d$-$4f$ Coulomb interaction.

Key words: resonant x-ray scattering; antiferroquadrupole order; DyB_2C_2

Resonant x-ray scattering (RXS) has recently attracted much interest as a useful tool to investigate the orbital order, which neutron scattering experiments are usually difficult to probe. A resonant enhancement has been found near the $L_{\rm III}$ edge on the antiferro-quadrupole (AFQ) ordering superlattice spots in DyB_2C_2 [1]. The main peak at $\hbar\omega = 7792$ eV is assigned to come from the E_1 process. Therefore, $5d$ states of Dy in the intermediate state are to be modulated in accordance with the superlattice spots. In this paper, we calculate the RXS spectra on the basis of a microscopic model that $4f$ states are atomic while $5d$ states form a band with an appropriate density of states (DOS). We have already carried out a similar calculation in CeB_6 [2].

The DyB_2C_2 crystal takes a tetragonal form at high temperatures (see Fig. 1). Dy^{3+} ion is approximately in the $4f^9$-configuration ($^6H_{15/2}$). The degeneracy may be lifted by the tetragonal crystal field, which may be expressed within the subspace of $J = 15/2$ as

$$H_{\rm crys} = A\{3J_z^2 - J(J+1)\}$$
$$+ B[\frac{2}{7}\{35J_z^4 - 30J(J+1)J_z^2 + 25J_z^2$$
$$+3J^2(J+1)^2 - 6J(J+1)\} - J_+^4 - J_-^4]. \quad (1)$$

The ground state is a Kramers doublet, $|\pm\frac{1}{2}\rangle$, and the first excited state is also a doublet, $|\pm\frac{3}{2}\rangle$ for $B = 0$. The last term $-B(J_+^4 + J_-^4)$ may make the states of $|M| > \frac{3}{2}$ admix to those of $M = \pm\frac{1}{2}$ and $\pm\frac{3}{2}$. ($|M\rangle$ represents the state of $J_z = M$). According to the specific heat measurement, the entropy per Dy atom is found $\sim \ln 4$ for $T > T_Q$ ($= 24.7$ K), indicating that the lowest four states are nearly degenerate. That situation can be realized by putting $B/A = 0.0029$.

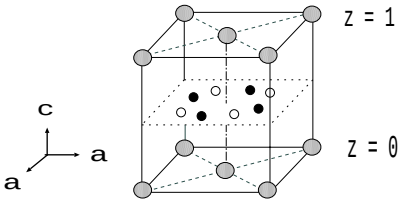

Fig. 1. Sketch of the crystal structure of DyB_2C_2 ($P4/mbm$: $a = 5.341$ Å, $c = 3.547$ Å at 30 K). Gray large circles are Dy atoms. Solid and open small circles are B and C atoms, respectively.

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The RXS spectra are calculated in the AFQ phase of the O_{xy}-type ($O_{xy} \equiv \frac{\sqrt{3}}{2}(J_xJ_y + J_yJ_x)$) with the ordering vector $\mathbf{Q} = (0, 0, \pi/c)$. Assuming the conventional RXS geometry, we use the expression,

$$\frac{d\sigma}{d\Omega}\Big|_{\mu\to\mu'} \propto |\sum_{\alpha\alpha'} P_{\alpha}^{\prime\mu'} M_{\alpha\alpha'}(\mathbf{G},\omega) P_{\alpha'}^{\mu}|^2, \tag{2}$$

with

$$M_{\alpha\alpha'}(\mathbf{G},\omega) = \frac{1}{\sqrt{N}} \sum_j \sum_{n,\Lambda} p_n(j) \exp(-i\mathbf{G}\cdot\mathbf{r}_j) \times \frac{\langle\psi_n(j)|x_\alpha(j)|\Lambda\rangle\langle\Lambda|x_{\alpha'}(j)|\psi_n(j)\rangle}{\hbar\omega - (E_\Lambda - E_n(j)) + i\Gamma}, \tag{3}$$

where j runs over Dy lattice sites. Here $\hbar\omega$ is the photon energy, and $\mathbf{G}$ is the scattering vector. The P^μ and P'^μ are geometrical factors for the incident and scattered photons (see Ref. [2]). The life-time broadening width Γ of the core hole is assumed to be 2.5 eV. The $x_\alpha(j)$'s are dipole operators.

The $|\psi_n(j)\rangle$ represents the initial state with energy $E_n(j)$ and the probability $p_n(j)$. In the AFQ phase, we specify $|\psi_1(j)\rangle$ and $|\psi_2(j)\rangle$ by the eigenfunctions of O_{xy} with a positive eigenvalue (doubly degenerate) while $|\psi_3(j)\rangle$ and $|\psi_4(j)\rangle$ by those with a negative eigenvalue. As an extreme case of quadrupole polarization, we put $p_1(j) = p_2(j) = \frac{1}{2}$, $p_3(j) = p_4(j) = 0$ for site j on the A sublattice, and $p_1(j) = p_2(j) = 0$, $p_3(j) = p_4(j) = \frac{1}{2}$ for site j on the B sublattice. Note that the magnetic moment vanishes if $|\psi_1\rangle$ and $|\psi_2\rangle$ ($|\psi_3\rangle$ and $|\psi_4\rangle$) are equally populated at A(B) sublattice.

The intermediate state $|\Lambda\rangle$ consists of an excited electron on $5d$ states and a hole on $2p$ states with energy E_Λ. The core-hole potential makes the $4f$ states different from those in the initial state. We consider only the states of $J = 15/2$ in the $4f^9$-configuration, taking full account of $5d$-$4f$, $5d$-$2p$, $4f$-$2p$ Coulomb interactions. We use the atomic values for the Slater integrals reduced by 20% (much reduced values for F^0). The $5d$ DOS is assumed to be the same as that in CeB_6, and the band effect is treated by introducing the local Green's function. See Ref. 2 for details of the calculation procedure.

Figure 2 shows the calculated RXS spectra as a function of photon energy at the azimuthal angle $\psi = 45^\circ$ for $\mathbf{G} = (00\frac{5}{2})$. The core hole energy is adjusted such that the main peak energy coincides with the experimental one. We obtain sufficient intensities without taking account of the lattice distortion. The single peak structure agrees with the recent experiment (The pre-edge peak from the E_2 process is not discussed here.).

Figure 3 shows the intensity of the main peak as a function of the azimuthal angle for $\mathbf{G} = (00\frac{5}{2})$. The intensity for the $\sigma \to \pi'$ channel shifts by 45° from the position of the $\sigma \to \sigma'$ channel. Their relative intensities agree well with the experiment. If the eigenfunctions of $O_{x^2-y^2}$ is used in the initial state, the azimuthal angle dependence will shift by 45° from the positions for O_{xy}. Therefore, the azimuthal dependence is quite useful to determine the type of quadrupole order.

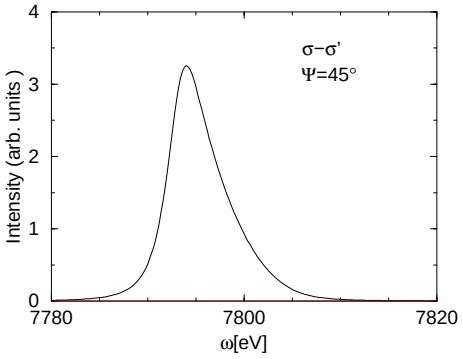

Fig. 2. Calculated RXS spectra for $\mathbf{G} = (00\frac{5}{2})$ at $\psi = 45^\circ$, as a function of photon energy. The intensity for the $\sigma \to \pi'$ channel vanishes.

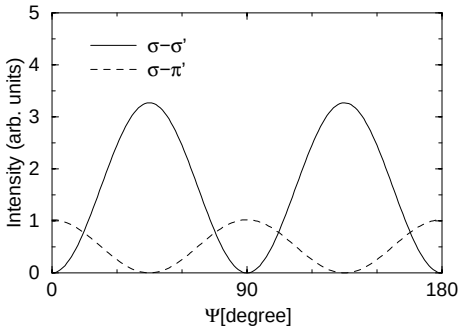

Fig. 3. Azimuthal angle dependence of the RXS intensity of the peak at $\hbar\omega = 7792$ eV for $\mathbf{G} = (00\frac{5}{2})$.

In summary, we have calculated the RXS spectra in DyB_2C_2. Without any lattice distortion, we have obtained sufficient intensities and the azimuthal angle dependence, in good agreement with the experiment. This result suggests that the RXS intensity comes mainly from the anisotropic part of the $5d$-$4f$ Coulomb interaction. This is completely opposite to the K edge RXS in transition-metal compounds, where the intensity comes mainly from the lattice distortion [3].

References

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