A theory of the electric quadrupole contribution to resonant x-ray scattering:
Application to multipole ordering phases in Ce$_{1-x}$La$_x$B$_6$

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Resonant x-ray scattering (RXS) has recently attracted much attention, since strong x-ray intensities have become available from the synchrotron radiation. It is described by a second-order process that a core electron is excited into unoccupied states by absorbing incident x-rays and that electron is recombined with the core hole by emitting x-rays. The RXS has been recognized as a useful probe to investigate spatially varying multipole orderings, which the conventional neutron scattering is usually difficult to detect.

For probing the spacial variation of order parameters, x-ray wavelengths need to be order of the variation period. In transition metals, the $K$ period. In transition metals, the $K$ edge, the possibility of the orbital ordering has already been explored in transition-metal compounds. The RXS intensities are observed at superlattice Bragg spots, which are interpreted as originat-ting from the modulation in the 4$p$ band, since the process involves the excitation of a 1$s$ electron to unoccupied 4$p$ states.

Because the ordering pattern is usually controlled by electrons in the 3$d$ band, the mechanism which causes the modulation is not necessarily trivial. Actually, for most of transition-metal compounds, both experimental studies and theoretical studies based on electronic structure calculations have revealed that the RXS intensities are brought about by the hybridization between the 4$p$ band and the 2$p$ band of the neighboring anions rather than the direct Coulomb interaction between the electron in the 4$p$ band and electrons in the 3$d$ band. This result is reasonable because of the extended nature of the 4$p$ state.

On rare earth metal compounds such as CeB$_6$, DyB$_2$C$_2$, the $L_{2,3}$ edges in the $E1$ transition are used because of the requirement for x-ray wavelength. The RXS spectra in the $E1$ transition from the antiferroquadrupole (AFQ) phase of CeB$_6$ were studied both experimentally and theoretically. Although the experiments and the theory give sufficiently consistent results, the relation to the multipole orderings which 4$f$ electrons mainly involve is rather indirect, since the resonance is caused by the excitation of a 2$p$ electron to 5$d$ states. This shortcoming may be overcome by using the quadrupole (E2) transition at the $L_{2,3}$ edges, where a 2$p$ electron is promoted to partially filled 4$f$ states. Using the E2 transition has another merit that octupole and hexadecapole orderings are directly detectable. This contrasts with the $E1$ transition, where only dipole and quadrupole orderings are detectable. Of course, intensities in the E2 transition are usually much smaller than those in the E1 transition.

In this paper, we derive a general formula of the RXS amplitudes in the localized electron picture, in which the electronic structure at each atom is assumed to be well described by an atomic wavefunction under the crystal electric field (CEF). Historically, the research in such a direction was started by using the framework borrowed from resonant $\gamma$-ray scattering. Starting from the works by Blume and Gibbs and by Hannon et al.,

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the form of the RXS amplitude had been investigated in several works. The RXS amplitude can be summarized into an elegant form by using vector spherical harmonics. Unfortunately, it has little practical usage because it is difficult to deduce meaningful information when there is no restriction on the intermediate state of the scattering process. A widely-adopted approximation for practical use is the so-called "fast collision (FC) approximation". This replaces the intermediate state energy in the energy denominator of the RXS amplitude by an averaged value, allowing the denominator out of the summation. Thereby, the multiplet splitting of the intermediate state is neglected, leading to an assumed form (usually a Lorentzian form) for the energy profile.

However, recent experiments show deviation from the Lorentzian form in several materials. We improve the situation by taking the energy dependence of the intermediate state under the assumption that the intermediate Hamiltonian describing the scattering process preserves spherical symmetry. This assumption is justified when the CEF energy and the intersite interaction are much smaller than the multiplet energy in the intermediate state as is expected in many localized electron materials. We have already reported the formula for the E1 transition, having successfully applied to the analysis of the E1 RXS spectra in URu2Si2 and NpO2. This paper is an extension of those works to the E2 transition. The obtained formula makes it possible to analyze the energy profiles of the spectra in contrast with the FC approximation. In addition, the formula is suitable to discuss the relation of the RXS spectra to multipole order parameters because it is expressed by means of the expectation values of the multipole order parameters.

We demonstrate the usefulness of the formula by calculating the RXS spectra in multipole ordering phases of Ce1−xLaxB6. First, we investigate the E2 RXS spectra at the Ce L2,3 edges from the AFQ ordering phase (phase II) in the non-diluted material CeB6. Analysis utilizing our formula reveals that the E2 RXS spectra in phase II consist of a mixture of the quadrupole and hexadecapole energy profiles. The calculated intensities suggest the possibility that the E2 signal at the Ce L2,3 edges can be detectable in this material.

For the intermediate doping range $x \approx 0.3 \sim 0.5$, Ce1−xLaxB6 falls into a new phase (phase IV) whose origin of this discrepancy is still unanswered. It will be pointed out that the RXS peak intensity from the AFQ phase concomitant with the induced hexadecapole contribution also gives rise to the same symmetry of the $\psi$-dependence as obtained from the AFO phase. Thus, although the AFO order is plausible in many respects, it seems difficult to rule out the AFQ order on the basis of the azimuthal angle dependence alone. In addition, we calculate the energy dependence of the RXS spectra at the Ce L2,3 edges. Assuming both the AFO and AFQ orders, we obtain the spectral shapes at the L2 edge, which agree with the experimental one for Ce0.7La0.3B6. On the other hand, the spectral shapes at the L3 edge are found slightly different between two phases, with intensities the same order of magnitude of the reported one at the L2 edge.

The present paper is organized as follows. A general expression for RXS amplitudes is obtained in Sec. II. Analysis of RXS spectra in Ce1−xLaxB6 are presented in Sec. III. Section IV is devoted to concluding remarks. In Appendix, we show several expressions required to obtain the RXS amplitude formula.

II. THEORETICAL FRAMEWORK OF RXS

A. A second-order optical process

The RXS is described by a second-order optical process, where a core electron is excited to unoccupied states by absorbing x-rays and that electron is recombined with the core hole by emitting x-rays. Since the wavefunction of core electron is well localized, the RXS amplitude may be given by a sum of contributions from individual ions. Using a geometrical arrangement shown in Fig. 1, we express the RXS amplitude $f(\epsilon, \epsilon', \mathbf{k}, \mathbf{k}', \omega)$ for the incident x-ray with momentum $\mathbf{k}$, polarization $\epsilon$, and the scattered x-ray with momentum $\mathbf{k}'$, polarization $\epsilon'$ as

\[
f(\epsilon, \epsilon', \mathbf{k}, \mathbf{k}', \omega) = \sum_{n=1} f^{(n)}(\epsilon, \epsilon', \mathbf{k}, \mathbf{k}', \omega), \quad (2.1)
\]

\[
f^{(n)}(\epsilon, \epsilon', \mathbf{k}, \mathbf{k}', \omega) \propto \frac{1}{\sqrt{N}} \sum_{j} e^{-i \mathbf{G}_{\epsilon} \cdot \mathbf{r}_{j}} \times M^{(n)}_{j}(\epsilon, \epsilon', \mathbf{k}, \mathbf{k}', \omega), \quad (2.2)
\]

where $n$ is the order of multipole component, $\mathbf{G}_{\epsilon}$ is the wavevector of the core electron, and $M^{(n)}_{j}(\epsilon, \epsilon', \mathbf{k}, \mathbf{k}', \omega)$ are the matrix elements for RXS.
Note that the quadrupole operators ˜z polarization where G = ˜z = 0.

The (112) surface is in the scattering plane at ψ = 0.

with

\[ M_j^{(1)}(\epsilon, \epsilon', \omega) = \sum_{\mu, \mu'} \epsilon'_\mu \epsilon_{\mu'} \times \sum_{\Lambda} \frac{\langle \psi_0 | x_{\mu,j} | \Lambda \rangle \langle \Lambda | x_{\mu',j} | \psi_0 \rangle}{\hbar c - (E_\Lambda - E_0) + i\Gamma} \]

\[ M_j^{(2)}(\epsilon, \epsilon', k, k', \omega) = \frac{k^2}{9} \sum_{\mu, \mu'} q_{\mu'}(\mathbf{k}', \epsilon') q_{\mu'}(\mathbf{k}, \epsilon) \times \sum_{\Lambda} \frac{\langle \psi_0 | \tilde{z}_{\mu,j} | \Lambda \rangle \langle \Lambda | \tilde{z}_{\mu',j} | \psi_0 \rangle}{\hbar c - (E_\Lambda - E_0) + i\Gamma} \]

where G (= k' - k) is the scattering vector, and N is the number of sites (j)'s. The |ψ0⟩ represents the ground state with energy E0, while |Λ⟩ represents the intermediate state with energy E_Λ. The Γ describes the life-time broadening width of the core hole. Equation (2.3) describes the E1 transition, where the dipole operators x_{µ,j}'s are defined as x_{1,j} = x_j, x_{2,j} = y_j, and x_{3,j} = z_j in the coordinate frame fixed to the crystal axes with the origin located at the center of site j. Equation (2.4) describes the E2 transition, where the quadrupole operators are defined by \( \tilde{z}_{1,j} = \frac{1}{2} (x_j^2 - y_j^2) \), \( \tilde{z}_{2,j} = \frac{1}{2} (3x_j^2 - r_j^2) \), \( \tilde{z}_{3,j} = \sqrt{3} y_j x_j \), \( \tilde{z}_{4,j} = \sqrt{3} z_j x_j \) and \( \tilde{z}_{5,j} = \sqrt{3} x_j y_j \). Factors \( q_{\mu}(\mathbf{k}, \epsilon) \) and \( q_{\mu'}(\mathbf{k}', \epsilon') \) with \( \mathbf{k} = k/|k| \) and \( \mathbf{k}' = k'/|k'| \) are defined as a second-rank tensor,

\[ q_{\mu}(A, B) = \begin{cases} \frac{\sqrt{3}}{2} (A_x B_x - A_y B_y) & \text{for } \mu = 1, \\ \frac{3}{2} (A_x B_x - A_y B_y) & \text{for } \mu = 2, \\ \frac{\sqrt{3}}{2} (A_y B_x + A_z B_y) & \text{for } \mu = 3, \\ \frac{\sqrt{3}}{2} (A_z B_x + A_y B_z) & \text{for } \mu = 4, \\ \frac{\sqrt{3}}{2} (A_x B_y + A_z B_y) & \text{for } \mu = 5. \end{cases} \]  

(2.5)

Note that the quadrupole operators \( \tilde{z}_{\mu,j} \) are expressed as \( \tilde{z}_{\mu,j} = q_{\mu}(r_j, r_j) \).

\[ |\psi_0\rangle_j = \sum_{m} c_j(m)|J, m\rangle. \]  

(2.6)

In the intermediate state, however, the CEF and the intersite interaction may be neglected in a good approximation, since their magnitudes of energy are much smaller than those of the intra-atomic Coulomb interaction and the spin-orbit interaction (SOI) which give rise to the multiplet structure. Thus the Hamiltonian describing the intermediate state is approximated as preserving the spherical symmetry. In such a circumstance, the intermediate states are characterized by the total angular momentum at the core-hole site, that is, |Λ⟩ = |J', M, i⟩ with the magnitude J' and the magnetic quantum number M. The corresponding energy is denoted by \( E_{J', i} \), where we introduce the index i in order to distinguish multiplets having the same J' value but having different energy.

In the following, we discuss only on the E2 transition (Eq. (2.4)), because the E1 transition has been fully analyzed in our previous paper. First, we rewrite Eq. (2.4) as

\[ M_j^{(2)}(\epsilon, \epsilon', \mathbf{k}, \mathbf{k}', \omega) = \frac{k^2}{9} \sum_{\mu, \mu'} q_{\mu}(\mathbf{k}', \epsilon') q_{\mu'}(\mathbf{k}, \epsilon) M_j^{(2)}(\omega, j), \]  

(2.7)

\[ M_{\mu\mu'}(\omega, j) = \sum_{J', M, i} E_i(\omega, J') \langle \psi_0 | \tilde{z}_{\mu,j} | J', M, i \rangle \times \langle J', M, i | \tilde{z}_{\mu',j} | \psi_0 \rangle, \]  

(2.8)

with

\[ E_i(\omega, J') = \frac{1}{\hbar c - (E_{J', i} - E_0) + i\Gamma}. \]  

(2.9)

Then, inserting Eq. (2.6) for the ground state into Eq. (2.8), we obtain

\[ M_{\mu\mu'}^{(2)}(J, \omega) = \sum_{m, m'} c_j^*(m) c_j(m') M_{\mu\mu'}^{(2)m, m'}(\omega), \]  

(2.10)

with

\[ M_{\mu\mu'}^{(2)m, m'}(\omega) = \sum_{J'} \sum_{i=1}^{N_{J'}} E_i(\omega, J') \times \sum_{M=-J'}^{J} \langle J, m | \tilde{z}_{\mu,j} | J', M, i \rangle \langle J', M, i | \tilde{z}_{\mu',j} | m', m' \rangle. \]  

(2.11)
We have suppressed the index $j$ specifying the core-hole site. The number of the multiplets having the value $J'$ is denoted by $N_{J'p}$. The selection rule for the $E2$ transition confines the range of the summation over $J'$ to $J' = J,J + 1,J + 2$.

Now we analyze the matrix element of the type $\langle J, m|\tilde{s}_\mu J', M \rangle$ by utilizing the Wigner-Eckart (WE) theorem for a tensor operator

$$\langle J, m|s_\mu J' M \rangle = (-1)^{J+m-2}\sqrt{2J+1} \times \left( \begin{array}{c} J \mu' \mu \nu \end{array} \right) (J|V_2||J') \quad (2.12)$$

with $s_{\pm 2} = (\tilde{z}_1 \pm i\tilde{z}_3)/\sqrt{2}$, $s_{\pm 1} = \mp(\tilde{z}_4 \pm i\tilde{z}_3)/\sqrt{2}$ and $s_0 = \tilde{z}_2$. The symbol $(J|V_2||J')$ denotes the reduced matrix element of the set of irreducible tensor operator of the second rank. Because of the nature of the quadrupole operators, a condition $|m-m'| \leq 4$ has to be satisfied for non-vanishing $M^{(2)}_{\mu \mu'} (\omega)$. After a straightforward but tedious calculation with the help of the WE theorem, we obtain non-zero $M^{(2)}_{\mu \mu'} (\omega)$'s. Then, we perform the summation over $m$ and $m'$ in Eq. (2.10). The result is summarized by introducing the expectation values of the components of the multipole operators as follows:

$$M^{(2)}_{\mu \mu'} (j, \omega) = \sum_{\lambda=1}^{4} \alpha^{(\nu)}_{E2} (\omega) \sum_{\lambda=1}^{2\nu+1} [M^{(\nu)}_{\lambda} ]_{\mu,\mu'} \langle \psi_0 | z_\nu^{(\nu)} | \psi_0 \rangle,$$

(2.13)

where the $\lambda$th component of rank $\nu$ tensor $z^{(\nu)}_\lambda$ in real basis $(1 \leq \lambda \leq 2\nu + 1)$ is defined in Table I. The $z^{(\nu)}_\lambda$ is constructed from the irreducible spherical tensor $T^{(n)}_\lambda$ through the unitary transformation $U^{(\nu)}$. The definitions of $T^{(n)}_\lambda$ and $U^{(\nu)}$ as well as the energy profile $\alpha^{(\nu)}_{E2} (\omega)$ are given in Appendix A. The matrix element of $M^{(\nu)}_{\lambda}$ is expressed as

$$[M^{(\nu)}_{\lambda} ]_{\mu,\mu'} = \frac{(-\nu)^\nu}{(2|T_{\nu}|2)!} \sqrt{\frac{2\nu + 1}{5}} \sum_{\ell = -\nu}^{\nu} \sum_{\ell' = -\nu}^{\nu} U^{(2)}_{\mu \ell} \times \sum_{n=-\nu}^{\nu} U^{(\nu)}_{n \lambda} ([T^{(\nu)}_n]_{\ell \ell'}) [U^{(2)}_{\ell \ell'}]_{\mu' \nu},$$

$$= \frac{(-\nu)^\nu}{(2|T_{\nu}|2)!} \sqrt{\frac{2\nu + 1}{5}} \sum_{\ell = -\nu}^{\nu} \sum_{\ell' = -\nu}^{\nu} (-\nu)^\nu U^{(2)}_{\mu \ell} \times \sum_{n=-\nu}^{\nu} U^{(\nu)}_{n \lambda} \left( \frac{2 \nu n - \ell}{n - \ell} \right) [U^{(2)}_{\ell \ell'}]_{\mu' \nu'},$$

(2.14)

with

$$(2|T_{\nu}|2)! = \frac{1}{2\nu!} \sqrt{\frac{5 + \nu}{5(4 - \nu)}},$$

(2.15)

Finally, inserting Eq. (2.13) into Eq. (2.10) and using

| Table I: Definition of the operator equivalence of the multipole order components. The overline denotes the symmetrization, for instance, \( \overline{X^2 Y} = X^2 Y + X Y X + Y X^2 \). |
|-----------------|-----------------|-----------------|
| \( z^{(1)}_1 \) | \( J_1 \) | \( J_1 \) |
| \( z^{(1)}_2 \) | \( J_2 \) | \( J_2 \) |
| \( z^{(2)}_1 \) | \( J_3 \) | \( J_3 \) |
| \( z^{(2)}_2 \) | \( J_4 \) | \( J_4 \) |
| \( z^{(3)}_1 \) | \( J_5 \) | \( J_5 \) |
| \( z^{(3)}_2 \) | \( J_6 \) | \( J_6 \) |
| \( z^{(4)}_1 \) | \( J_7 \) | \( J_7 \) |
| \( z^{(4)}_2 \) | \( J_8 \) | \( J_8 \) |

Eq. (2.14), we obtain the final expression,

$$M^{(2)}_{\mu \mu'} (e,e',k,k',\omega) = \frac{k^2}{9} \sum_{\nu=0}^{4} \alpha^{(\nu)}_{E2} (\omega) \sum_{\lambda=1}^{2\nu+1} P^{(\nu)}_{\lambda} (e,e',k,k') \langle \psi_0 | z^{(\nu)}_\lambda | \psi_0 \rangle,$$

(2.16)

where $P^{(\nu)}_{\lambda}$ are the geometrical factors defined as

$$P^{(\nu)}_{\lambda} (e,e',k,k') = \sqrt{2\nu + 1} \sum_{n=-\nu}^{\nu} (-\nu)^n P^{(\nu)}_{\lambda n} \sum_{m=-\nu}^{\nu} \left( \frac{2 \nu m - n}{m - n} \right) \times q_{m,n}(e',k).$$

(2.17)

Those for $\nu = 0, 1$ and 2 are expressed as relatively
simple forms:

\[ P^{(0)}(e, e', k, k') = \frac{1}{\sqrt{5}} \left[ (k' \cdot \hat{k})(e' \cdot e) + (k' \cdot \hat{e})(e' \cdot \hat{k}) \right], \]

(2.18)

\[ P^{(1)}(e, e', \hat{k}, \hat{k'}) = -\frac{i}{\sqrt{10}} \left[ (e' \cdot e)(\hat{k} \times \hat{e}) + (e' \cdot \hat{k})(\hat{e} \times e) \right] \\
+ (k' \cdot \hat{e})(e' \cdot e) + (e' \cdot \hat{k})(\hat{e} \cdot e)' \right], \]

(2.19)

\[ P^{(2)}(e, e', \hat{k}, \hat{k'}) = -\frac{3}{2\sqrt{14}} \left[ (e' \cdot e)\mu\mu(\hat{k} \times \hat{e}, e') + (\hat{e} \cdot e)\mu\mu(\hat{k} \times k', e') \right] \] .

(2.20)

For \( \nu = 1 \), indices \( \mu = 1, 2 \) and 3 serve as the Cartesian components \( x, y \) and \( z \), respectively. The corresponding expression of \( P^{(\nu)} \)’s for \( \nu = 3, 4 \) have complicated forms, which are summarized in Appendix B.

An expression similar to Eq. (2.16) has been derived by the FC approximation.\textsuperscript{14,15,16,17} However, this scheme has to put by hand the energy dependence. The present theory gives an explicit expression of the energy dependence, which is separated from the factor relating to the order parameter. Thus, the choice of the CEF parameters in the ground state does not affect the shape of energy profiles \( \alpha_{E2}^{(\nu)}(\omega) \).

III. APPLICATION TO MULTIPOLAR ORDERING PHASES IN Ce\textsubscript{1-x}La\textsubscript{x}B\textsubscript{6}

In this section, we demonstrate the usefulness of Eq. (2.16) by analyzing the RXS spectra in the \( E2 \) transition at the Ce \( L2,3 \) edges from Ce\textsubscript{1-x}La\textsubscript{x}B\textsubscript{6}.

A. Phase II in CeB\textsubscript{6}

The parent material CeB\textsubscript{6} experiences two-step phase transitions. It undergoes the first transition from paramagnetic (phase I) to an AFQ state (phase II) at \( T_Q = 3.2 \) K and the second transition to an antiferromagnetic (AFM) state (phase III) at \( T_N = 2.4 \) K under no external magnetic field. The AFQ order is known to be a Néel-type with a propagating vector \( \mathbf{Q}_0 = (\frac{2 \pi}{5}, \frac{2 \pi}{5}, \frac{2 \pi}{5}) \).

These phase transitions have been theoretically studied in a localized electron scheme, where each Ce ion is assumed to be trivalent in the \( 4f^1 \) configuration. Its ground multiplet is a \( \Gamma_8 \) quartet confined within the \( J = \frac{3}{2} \) subspace under the cubic symmetry. Using states \( \{|J_z = m\rangle\} \), the four bases \( |\pm, \sigma \rangle \) (\( \sigma = \uparrow, \downarrow \)) may be expressed as

\[ |+\uparrow\rangle = \sqrt{\frac{5}{6}} |\uparrow\rangle + \frac{1}{2} |\downarrow\rangle, \]

(3.1)

\[ |-\downarrow\rangle = \frac{1}{2} |\uparrow\rangle, \]

(3.2)

and \( |\pm, \downarrow\rangle \) by replacing \( |m\rangle \) with \( | - m \rangle \). The intersite interaction may lift the fourfold degeneracy, leading to multipole orderings. Shihina et al. have derived such interaction from a microscopic model and obtained the phase diagram in agreement with experiments.\textsuperscript{30,31}

Instead of pursuing this direction, we simply assume the ordering pattern, and calculate the RXS spectra. The assumed ordering pattern selects a particular energy profile according to Eq. (2.16). Note that the quartet \( \Gamma_8 \) consists of 16 degrees of freedom, which are exhausted by three components of dipole, five components of quadrupole and seven components of octupole operators as well as an identical operator. Thereby the hexadecapole operators \( H^q_x, H^q_y, H^q_z, H^s_x, H^s_y, H^s_z \) and \( H^s_r \) are equivalent to identical operator, \( O_{x2-y2}, O_{z2-x2}, O_{yz}, O_{zx} \) and \( O_{xy} \), respectively, while \( H^s_{xy}, H^s_{yz}, H^s_{zx} = 0 \). Therefore, as long as a contribution from \( \alpha_{E2}^{(2)}(\omega) \) exists, that from \( \alpha_{E2}^{(4)}(\omega) \) automatically exists.

The order parameter in phase II is believed to be the \( O_{xy} \)-type. Operator \( O_{xy} \) has two degenerate eigenstates of eigenvalue \(-1\) and two degenerate eigenstates of eigenvalue \(+1\), that is,

\[ O_{xy} = \frac{1}{\sqrt{2}} \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \]

(3.3)

within the bases of eigenfunctions. The AFQ phase may be constructed by assigning two degenerate eigenstates with eigenvalue \(-1\) to one sublattice and those with eigenvalue \(+1\) to the other sublattice. The degeneracy of the Kramers doublet would be lifted in the AFM phase with further reducing temperatures. Within the same bases in order, typical dipole and octupole operators are represented by

\[ J_z = \begin{pmatrix} \frac{-2}{\sqrt{5}} & 0 & 0 & \frac{-2}{\sqrt{5}} \\ 0 & \frac{2}{\sqrt{5}} & \frac{-2}{\sqrt{5}} & 0 \\ 0 & \frac{2}{\sqrt{5}} & \frac{-2}{\sqrt{5}} & 0 \\ \frac{2}{\sqrt{5}} & 0 & 0 & \frac{-2}{\sqrt{5}} \end{pmatrix}, \]

(3.4)

\[ T^y_z = \begin{pmatrix} 0 & 0 & 0 & i3\sqrt{5} \\ 0 & 0 & -i3\sqrt{5} & 0 \\ 0 & i3\sqrt{5} & 0 & 0 \\ -i3\sqrt{5} & 0 & 0 & 0 \end{pmatrix}. \]

(3.5)

These forms indicate that the \( O_{xy} \) order could accompany neither the \( J_z \) order nor the \( T^y_z \) order. Therefore, the \( O_{xy} \) order selects the energy profiles \( \alpha_{E2}^{(2)}(\omega) \) and \( \alpha_{E2}^{(4)}(\omega) \) according to Eq. (2.16).

In the actual calculation of energy profile, we take into account full Coulomb interactions between \( 2p \) and \( 4f \) electrons, between \( 2p \) electrons, and between \( 4f \) electrons in the configuration \((2p)^6(4f)^2\). The spin-orbit interaction (SOI) of \( 2p \) and \( 4f \) electrons are considered too. The Slater integrals necessary for the Coulomb interactions and the SOI parameters are evaluated within the Hartree-Fock approximation.\textsuperscript{32,33}
over the contributions from three domains is performed. It depends strongly on domains. An incoherent addition of the scattering vector \( \mathbf{G} \) dependent of \( \nu \) the term of \( \sigma \) and some structures, which depend on the \( \Gamma \) value. It may be appropriate to use \( \Gamma = 2.0 \) eV. Three domains are assumed to have equal volumes. The solid and broken lines represent the \( \sigma-\sigma' \) and \( \sigma-\pi' \) channels, respectively.

Figure 2 shows the RXS spectra as a function of photon energy, calculated with the core-hole lifetime width \( \Gamma = 2.0 \) eV and 1.0 eV. The energy of the Ce 2p-core level is chosen such that the peak of the RXS spectra at the Ce L\(_3\) edge coincides with the experiment for CeB\(_6\). We find that the absolute value of \( \alpha_E^{(3)}(\omega) \) is much smaller than that of \( \alpha_E^{(3)}(\omega) \). However, the smallness is compensated by a large value of \( \langle \psi_0 | z_{\text{A}}^{(4)} | \psi_0 \rangle \), and thereby both terms contribute to the intensity. The calculated spectra show asymmetry and some structures, which depend on the \( \Gamma \) value. When the \( O_{xy} \) order is realized, the \( O_{yz} \) and \( O_{zx} \) orders are also possible to be realized. In actual crystals, three orders may constitute domains, whose structure affects the azimuthal angle dependence of the RXS spectra. Figure 3 shows the peak intensity as a function of azimuthal angle at the Ce L\(_3\) edge from the AFQ phase (phase II). \( \Gamma = 2.0 \) eV. Three domains are assumed to have equal volumes. The solid and broken lines represent the \( \sigma-\sigma' \) and \( \sigma-\pi' \) channels, respectively.

Figure 2 shows the RXS spectra at the Ce L\(_3\) (top) and L\(_2\) (bottom) absorption edges from the AFQ phase (phase II). \( \Gamma = 2.0 \) eV and 1.0 eV, \( \mathbf{G} = (2,2,2) \), and \( \psi = 0 \). Only the spectra in the \( \sigma-\sigma' \) channel are displayed.

Figure 3 shows the peak intensity as a function of azimuthal angle at the Ce L\(_3\) edge from the AFQ phase (phase II). \( \Gamma = 2.0 \) eV. Three domains are assumed to have equal volumes. The solid and broken lines represent the \( \sigma-\sigma' \) and \( \sigma-\pi' \) channels, respectively.

In the specific heat curve suggests the existence of the long range order [46], no neutron scattering experiment has found an evidence of long range magnetic order [46,47]. It is suggested [38,39] that the AFO order characterizes phase IV, which is supported by the observation of the trigonal distortion [46]. Recently, Mannix et al. measured the RXS spectra at the L\(_2\) edge in the E2 transition in Ce\(_{0.7}\)La\(_{0.3}\)B\(_6\), claiming that the signal arises from the AFO order [49]. The analysis of the azimuthal angle dependence by Kusunose and Kuramoto supports the AFO order in phase IV [49]. However, there exists at least one prominent discrepancy between the experiment and the theory about the azimuthal angle dependence which we shall address later.

Keeping two possibilities, the quadrupole and octupole orders, for phase IV, we analyze the spectra on the basis of Eq. (2.10). Since the trigonal distortion is observed along the body-diagonal direction, we assume that the order parameter is of \( T_{111}^3 \) type \( (T_{111}^3 = (T_{111}^3 + T_{222}^3 + T_{333}^3)/\sqrt{3}) \) or \( O_{111} \) type \( (O_{111} = \sum_{\alpha=xy} (O_{\alpha x} + O_{\alpha z} + O_{2z})/\sqrt{3}) \). The \( T_{111}^3 \) type can be ruled out because this type carries a substantial antiferromagnetic moment, which is against the experimental finding. Since \( T_{111}^3, O_{111} = 0 \), both operators are simultaneously diagonalized. Within the bases of eigenfunctions, they are represented as

\[
T_{111}^3 = \begin{pmatrix}
-3\sqrt{10} & 0 & 0 \\
0 & 3\sqrt{10} & 0 \\
0 & 0 & 0
\end{pmatrix}, \quad (3.6)
\]

\[
O_{111} = \begin{pmatrix}
-1 & 0 & 0 \\
0 & -1 & 0 \\
0 & 0 & 1
\end{pmatrix}. \quad (3.7)
\]

B. Phase IV in Ce\(_{1-x}\)La\(_x\)B\(_6\)

The La diluted material Ce\(_{1-x}\)La\(_x\)B\(_6\) with \( x \approx 0.3 \sim 0.5 \) has an additional phase IV whose order parameter is not well understood yet [49]. Although a large discontinuity in the specific heat curve suggests the existence of the long range order [45], no neutron scattering experiment has found an evidence of long range magnetic order [46,47]. It is suggested [38,39] that the AFO order characterizes phase IV, which is supported by the observation of the trigonal distortion [46]. Recently, Mannix et al. measured the RXS spectra at the L\(_2\) edge in the E2 transition in Ce\(_{0.7}\)La\(_{0.3}\)B\(_6\), claiming that the signal arises from the AFO order [49]. The analysis of the azimuthal angle dependence by Kusunose and Kuramoto supports the AFO order in phase IV [49]. However, there exists at least one prominent discrepancy between the experiment and the theory about the azimuthal angle dependence which we shall address later.
Within the same bases, the dipole operator $J_{111}$ ($= (J_x + J_y + J_z)/\sqrt{3}$) is represented as

$$J_{111} = \begin{pmatrix} 0 & z_1 & 0 & 0 \\ z_1^* & 0 & 0 & 0 \\ 0 & 0 & -\frac{1}{\sqrt{3}} & 0 \\ 0 & 0 & 0 & \frac{1}{\sqrt{3}} \end{pmatrix}, \quad (3.8)$$

where $z_1 = \sqrt{\frac{3}{18}}(1 + i11\sqrt{2})$.

1. AFO order

The AFO order may be constructed by assigning the eigenstate of the $T_{111}^\beta$ with eigenvalue $-3\sqrt{10}$ to one sublattice and that with $3\sqrt{10}$ to the other sublattice. Then, the order parameter vector $(\langle T_1^\beta \rangle, \langle T_2^\beta \rangle, \langle T_3^\beta \rangle)$ is pointing to the (111) direction. Equation (3.8) indicates that the AFO order accompanies the (111) direction. Equation (3.7) indicates that the AFO order accompanies the experimental. Equation (3.7) indicates that the AFO order accompanies the ferroquadrupole order, not the AFQ order. Therefore, according to Eq. (3.7), the RXS energy dependence is purely characterized by $|\alpha_E^{(3)}(\omega)|^2$.

Figure 4 shows the calculated $|\alpha_E^{(3)}(\omega)|^2$ as functions of the incident photon energy $\omega$ at the Ce L2 and L3 absorption edges with $\Gamma = 2.0$ eV and 1.0 eV, in comparison with the experiment of Mannix et al. (the non-resonant contribution is subtracted from the data). In the calculation, we use the same Slater integrals and the SOI parameters as in phase II. The spectral shapes depend strongly on the absorption edge they are observed. In particular, the tail part of the spectra at the L3 edge is drastically different from that at the L2 edge. This fact might be helpful to identify the character of the ordering pattern if the spectrum at the L3 edge is experimentally available. Since the peak intensity at the L3 edge is about 20% of that at the L2 edge, it can be said that experimental observation has a legitimate chance at the former edge. The L2 spectral shape reproduces well the experimental one showing broad single peak structure with a hump in the high energy region.

The energy profile $|\alpha_E^{(3)}(\omega)|^2$ looks similar to the spectral shape in phase II (Fig. 4) for $\Gamma = 2$ eV. One difference is a dip found at the L3 edge in $|\alpha_E^{(3)}(\omega)|^2$, which is absent in Fig. (4). The results are consistent with the fact that the absorption edge is observed. In this case, the differences are emphasized around the tail part of the high energy region, because multiplet structures of the intermediate state are emphasized. Note that, although $|\alpha_E^{(3)}(\omega)|^2$ is about two order of magnitude smaller than $|\alpha_E^{(2)}(\omega)|^2$, the smallness is compensated by the large factor of $|\langle T_{111}^\beta \rangle|^2 \approx 0.29$, resulting in the same order of magnitude of the spectral intensity as in phase II (Fig. 4).

If the octupole order parameter vector $(\langle T_1^\beta \rangle, \langle T_2^\beta \rangle, \langle T_3^\beta \rangle)$ can point to the (111) direction, it is also possible to point to the (111), (1T1), and (1T1) directions. These four orders usually constitute domains. The azimuthal angle dependence is different for different domains, as shown in Figs. 5(a) and (b). If you collect the contributions from domains with equal weight, the maximum intensity in the $\sigma-\pi'$ channel becomes nearly equal to that in the $\sigma-\sigma'$ channel. The experimental data show the maximum intensity in the $\sigma-\pi'$ channel is about the half of that in the $\sigma-\sigma'$ channel, as shown in Fig. 5(c). This may be attributed to the slightly different setup for different polarizations and/or to the extrinsic background from the non-resonant contribution, as discussed by Kusunose and Kuramoto. They reduced the intensity in the $\sigma-\pi'$ channel by simply multiplying a factor 0.6. Another possibility is that domain volumes are different among the domains. Collecting up the contributions with ratio 3 : 1 : 1 : 1 from the (111), (1T1), (1T1), and (1T1) domains, we have the result similar to that simply multiplying a factor 0.6 to the intensity in the $\sigma-\pi'$ channel, as shown in Fig. 5(c).

Thus, the sixfold and threefold symmetries in the $\sigma-\sigma'$ and $\sigma-\pi'$ channels are well reproduced in comparison with the experiment.

2. AFQ order

The AFQ order may be constructed by assigning the eigenstates of $O_{111}$ with eigenvalue $-1$ to one sublattice
and those with +1 to the other sublattice. The AFQ order accompanies no AFO order. The difference from phase II is that the order parameter \((O_{zy}, O_{zx}, O_{xy})\) is pointing to the (111) direction. Therefore, the spectral shape as a function of energy is nearly the same as in phase II. The azimuthal angle dependence depends strongly on domains, which is shown in Figs. 6(a) and (b). The sixfold symmetry in the \(\sigma-\sigma'\) and the \(\sigma-\pi'\) channels, respectively, where the solid (D1), broken (D2), dotted (D3), and broken-dotted (D4) lines represent the peak intensity of the domains (111), (1\(\overline{1}\)T), (1\(\overline{1}\)T), and (1\(\overline{1}\)T) respectively. Panel (c) shows the intensities collecting the contributions from the domains (111), (1\(\overline{1}\)T), (1\(\overline{1}\)T), and (1\(\overline{1}\)T) with ratio 3:1:1:1. The solid and broken lines represent the intensities in the \(\sigma-\sigma'\) and \(\sigma-\pi'\) channels, respectively. Filled and open circles are the experimental data for \(\text{Ce}_{0.7}\text{La}_{0.3}\text{B}_6\) respectively. Panel (c) shows the result collecting the contributions from the domains with equal weight. The solid and broken lines represent the \(\sigma-\sigma'\) and \(\sigma-\pi'\) channels, respectively. The curve in the latter channel is multiplied by a factor 0.6. Filled and open circles are the experimental data for \(\text{Ce}_{0.7}\text{La}_{0.3}\text{B}_6\)

FIG. 6: Peak intensities at the Ce \(L_2\) edges in the AFQ phase, as functions of azimuthal angle. Panels (a) and (b) display the peak intensities in the \(\sigma-\sigma'\) and the \(\sigma-\pi'\) channels, respectively, where the solid (D1), broken (D2), dotted (D3), and broken-dotted (D4) lines represent the peak intensity of the domains (111), (1\(\overline{1}\)T), (1\(\overline{1}\)T), and (1\(\overline{1}\)T) respectively. Panel (c) shows the result collecting the contributions from the domains with equal weight. The solid and broken lines represent the \(\sigma-\sigma'\) and \(\sigma-\pi'\) channels, respectively. The curve in the latter channel is multiplied by a factor 0.6. Filled and open circles are the experimental data for \(\text{Ce}_{0.7}\text{La}_{0.3}\text{B}_6\)

and relative intensity between two channel may be attributed to the consequence of these subtraction process.

We now turn our attention to the energy dependence of the spectra. Owing to our formula Eq. (2.10), the spectral shapes from the AFQ phase with \(O_{111}\) type in phase IV are the same as those with \(O_{xy}\) type in phase II (Fig. 2). Therefore, the energy dependence at the \(L_2\) edge is similar to that obtained from the AFO phase. On the other hand, the energy dependence at the \(L_3\) edge from the AFQ phase is different from that obtained from the AFO phase, which may help the identification of the ordering pattern realized in this material.

IV. CONCLUDING REMARKS

We have derived a general formula of the RXS amplitude in the \(E_2\) transition. The derivation is based on the assumption that the Hamiltonian describing the intermediate state of the scattering process preserves the spherical symmetry. The obtained formula is applicable to many \(f\) electron systems where a localized scheme gives
a good description. Although similar formulae have already been obtained, the present formula has two prominent advantages. One is that it is able to calculate the energy profile of the RXS spectra, because our treatment is free from the fast collision approximation adopted in the previous works. The other is that it is conveniently applicable to the systems possessing multipole order parameters.

We have demonstrated the usefulness of the derived formula by calculating the $E2$ RXS spectra in Ce$_{1-x}$La$_x$Bi$_6$. Phase II is believed to be an AFQ order of $O_{2g}$ type, and our formula dictates that the energy dependence is given by a combination of $a_{E2}^{(2)}(\omega)$ and $a_{E2}^{(4)}(\omega)$. We have obtained the RXS intensity in the same order of intensity as obtained by assuming the AFO order. This suggests that the $E2$ signal is detectable from phase II, although only the $E1$ signal has been reported in phase II of CeBi$_6$. Subsequently, we have calculated the RXS spectra by assuming the $T^{(3)}_{111}$-type AFO order, in order to clarify the order parameter of phase IV. The energy dependence $|a_{E2}^{(3)}(\omega)|^2$ has been obtained at the $L_2$ edge in agreement with the experiment in Ce$_{0.37}$La$_{0.63}$Bi$_6$. Unfortunately this is not used to discriminate between the AFO and AFQ orders, because the spectral shapes are nearly the same in the two ordering phases. On the other hand, the spectral shape at the $L_3$ edge has been found slightly different from the $L_2$ edge, which might help the identification of the ordering pattern. For the azimuthal angle dependence, we have reproduced the sixfold and threefold symmetries by assuming the AFO order, in agreement with the previous theoretical analysis and the experiment. The intensity in the $\sigma$-$\pi'$ channel becomes nearly equal to that in the $\sigma$-$\sigma'$ channel with the equal volume for four domains, while in the experiment the intensity in the former channel is found nearly half of that in the latter. This discrepancy may be removed by assuming uneven volumes among four domains. We have also analyzed the azimuthal angle dependence by assuming the $O_{111}$-type AFQ order. It is found that the simultaneously induced hexadecapole order gives rise to the sixfold and threefold symmetries. Although the agreement with the experiment is quantitatively not good, it may be difficult to rule out the AFQ order from phase IV on the basis of the azimuthal angle dependence alone. Since it depends strongly on the domain distribution, experiments controlling the domain distribution, if possible, might be useful to clarify the situation.

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| rank | $n$ | $T_{\nu}^{(\nu)}$ |
|------|----|----------------|
| 1    | $\pm 1$ | $\pm \frac{\sqrt{2}}{2} J_{\pm}$ |
|      | $0$   | $J_z$          |
| 2    | $\pm 2$ | $\pm \frac{\sqrt{2}}{2} J_{\pm}$ |
|      | $\pm 1$ | $\pm \frac{1}{2} \sqrt{7} J_{\pm} (2 J_z \pm 1)$ |
|      | $0$   | $[3 J_z - J (J + 1)]$ |
| 3    | $\pm 3$ | $\pm \frac{\sqrt{2}}{2} J_{\pm}$ |
|      | $\pm 2$ | $\pm \frac{1}{2} \sqrt{7} J_{\pm} (2 J_z \pm 1)$ |
|      | $\pm 1$ | $\pm \frac{1}{4} \sqrt{35} J_z \pm 15 J_z - 3 J (J + 1) + 6$ |
|      | $0$   | $[5 J_z - 3 J (J + 1) J_z + J_z]$ |
| 4    | $\pm 4$ | $\pm \frac{\sqrt{2}}{2} J_{\pm}$ |
|      | $\pm 3$ | $\pm \frac{1}{2} \sqrt{7} J_{\pm} (2 J_z \pm 3)$ |
|      | $\pm 2$ | $\pm \frac{1}{2} \sqrt{7} J_{\pm} [7 J_z^2 \pm 14 J_z - J (J + 1) + 9]$ |
|      | $\pm 1$ | $\pm \frac{1}{4} \sqrt{35} J_z \pm 21 J_z^2 + 19 J_z - 6 J (J + 1) J_z + 3 J (J + 1) + 6$ |
|      | $0$   | $[35 J_z^4 - 90 J (J + 1) J_z^2 + 25 J_z^2 + 3 J^2 (J + 1)^2 - 6 J (J + 1)]$ |

**APPENDIX A: DEFINITIONS OF SOME QUANTITIES USED IN SEC. II**

Let us define irreducible tensor operator of rank $\nu$ with the spherical basis. The $n$-th component $(-\nu \leq n \leq \nu)$ $T_{\nu}^{(\nu)}$ is defined recurrently as

$$
T_{\nu}^{(\nu)} = (-)^{\nu} \frac{(2 \nu - 1)!!}{(2 \nu)!!} J_{\nu}^{(\nu)},
$$

(A1)

$$
[J_-, T_{\nu}^{(\nu)}] = \sqrt{(\nu + n)(\nu - n + 1)} T_{\nu}^{(\nu)}.
$$

(A2)

Expressions for $T_{\nu}^{(\nu)}$'s are listed in Table III up to rank four. We can find $(2 n + 1) \times (2 \nu + 1)$ unitary matrix which connects the tensor operator with the spherical component $T_{\lambda}^{(\nu)}$ and that with the Cartesian component $z_{\lambda}^{(\nu)}$ which satisfies

$$
z_{\lambda}^{(\nu)} = \sum_{n=\nu}^{\nu} U_{\lambda n}^{(\nu)} T_{\nu}^{(\nu)},
$$

(A3)

and inversely,

$$
T_{\nu}^{(\nu)} = \sum_{\lambda=1}^{2 \nu + 1} [U^{(\nu)}]_{\lambda n} z_{\lambda}^{(\nu)}.
$$

(A4)

Explicit form of $U^{(\nu)}$ is summarized in Table III.
The energy dependence is contained in the functions $F_{J'}(\omega)$ as
\[ F_{J'}(\omega) = 4C_{J-J'+2} \sqrt{(2J+1)(2J'+1)} \frac{(J+J'-2)!}{(J+J'+3)!} \sum_{i=1}^{N_J} E_i(\omega, J'), \tag{A10} \]
where $n_{C_m} = \frac{n!}{m!(n-m)!}$ represents combination.

**APPENDIX B: GEOMETRICAL FACTORS**

The geometrical factors $P_{\mu}^{(\nu)}$ for $\nu = 3$ and 4 in Eq. 2.10 have rather complicated forms. For $\nu = 3$, they are summarized as follows:

\[
P_{1}^{(3)} = i \frac{1}{3 \sqrt{2}} \left[ [k' \times k'] \cdot q(\epsilon', \epsilon) + [\epsilon' \times \epsilon] \cdot q(k', k) \right.
\]
\[\left. + [k' \cdot k'] \cdot q(\epsilon', k) + [\epsilon' \times k] \cdot q(k', \epsilon) \right], \tag{B1} \]
\[
P_{2}^{(3)} = i \frac{5}{2} \left[ [k' \times k]_{\mu} \epsilon'_{\mu} \epsilon_{\mu} + [\epsilon' \times \epsilon]_{\mu} \hat{k}'_{\mu} \hat{k}_{\mu} \right.
\]
\[\left. + [k' \times \epsilon]_{\mu} \epsilon'_{\mu} \hat{k}_{\mu} + [\epsilon' \times k]_{\mu} \hat{k}'_{\mu} \epsilon_{\mu} \right]
\[+ i \frac{2 \sqrt{10}}{3 \sqrt{2}} P_{\mu}^{(1)} \text{ for } \mu = 2, 3, \text{ and } 4, \tag{B2} \]
\[
P_{3}^{(3)} = i \frac{3}{4} \left[ [k' \times \hat{k}]_{\mu} \epsilon_{\mu} \sum_{\mu', \mu''=5}^{7} \epsilon_{\mu''} \epsilon_{\mu'''} \right.
\]
\[\left. + i \frac{2}{4} \left[ [\epsilon' \times \epsilon]_{\mu} \sum_{\mu', \mu''=5}^{7} \epsilon_{\mu''} \epsilon_{\mu'''} \left( \hat{k}'_{\mu'} \hat{k}_{\mu''} - \hat{k}'_{\mu''} \hat{k}_{\mu'} \right) \right] \tag{B3} \]
\[
\]
For \( \nu = 4 \), the results are as follows.

\[
P_1^{(4)} = \sqrt{\frac{2}{15}} \left[ 5(k_x'k_x\epsilon_x'\epsilon_x + k_y'k_y\epsilon_y'\epsilon_y + k_z'k_z\epsilon_z'\epsilon_z) - P_1^{(6)} \right],
\]

\[
P_2^{(4)} = \sqrt{\frac{2}{15}} \left[ 5(k_x'k_x\epsilon_x'\epsilon_x - k_y'k_y\epsilon_y'\epsilon_y) - 2\sqrt{\frac{2}{7}} \left[ |k'|k|q_1(\epsilon', \epsilon) + |\epsilon'|\epsilon q_1(k', \k) \right] + |k'|\epsilon q_2(k', \k) + |\epsilon'|\k q_2(k', \k) \right],
\]

\[
P_3^{(4)} = \sqrt{\frac{2}{15}} \left[ 5(k_x'k_x\epsilon_x'\epsilon_x + k_y'k_y\epsilon_y'\epsilon_y - 2k_z'k_z\epsilon_z'\epsilon_z) \right],
\]

\[
P_\mu^{(4)} = \frac{1}{2\sqrt{6}} q_{\mu+1}(k', \k) \sum_{\mu', \mu''=4}^6 \epsilon_{\mu\mu'}\epsilon_{\mu''}(\epsilon_{\mu'}\epsilon_{\mu'} - \epsilon_{\mu''}\epsilon_{\mu''})
+ \frac{1}{2\sqrt{6}} q_{\mu+1}(\epsilon', \epsilon) \sum_{\mu', \mu''=4}^6 \epsilon_{\mu\mu'}\epsilon_{\mu''}(\epsilon_{\mu'}\epsilon_{\mu'} - \epsilon_{\mu''}\epsilon_{\mu''})
+ \frac{1}{2\sqrt{6}} q_{\mu+1}(\k', \k) \sum_{\mu', \mu''=4}^6 \epsilon_{\mu\mu'}\epsilon_{\mu''}(\epsilon_{\mu'}\epsilon_{\mu'} - \epsilon_{\mu''}\epsilon_{\mu''})
+ \frac{1}{2\sqrt{6}} q_{\mu+1}(\k', \k) \sum_{\mu', \mu''=4}^6 \epsilon_{\mu\mu'}\epsilon_{\mu''}(\epsilon_{\mu'}\epsilon_{\mu'} - \epsilon_{\mu''}\epsilon_{\mu''})
\]

where indices 4, 5, and 6 in the summations in Eq. (B7) serve as \( x, y, \) and \( z \), respectively. Similarly, indices 7, 8, and 9 appeared in the brakets in Eq. (B8) serve as \( x, y, \) and \( z \), respectively.

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