PHYSICAL REVIEW VOLUME 171, NUMBER 3 15 JULY 1968

Polarization Dependence of Two-Photon Absorption in Solids*

TODD R. BADER AND ALBERT GOLD
Institute of Optics, University of Rochester, Rochester, New York 14627
(Received 21 February 1968)

The polarization dependence of the rate of two-photon absorption has been calculated for all 32 crystallographic point groups. The method used by Inoue and Toyozawa to compute the polarization dependence of transitions from a Γ^1 (identity representation) ground state to an excited state of symmetry Γ^α (any single-valued representation) is extended to include all allowed two-photon transitions between pairs of states, each transforming according to any representation of the point group, including the double-valued representations. The results, presented in tabular form, are applicable to the analysis of transitions at point defects, and of band-to-band and exciton transitions at the center of the Brillouin zone.

I. INTRODUCTION

THE advent of the technique of two-photon spectroscopy[1] has opened the way to the use of two-photon absorption as a tool in the study of the symmetry of electronic states in solids. This method serves as a complement and supplement to conventional solid-state spectroscopy, particularly in exploring states not accessible in single-photon absorption.

One of the assets of two-photon spectroscopy is the wealth of information which may be obtained by varying the polarizations of the two beams with respect to one another and the crystal axes. Hence, it becomes useful to calculate and tabulate the form of these angular dependences so they may be readily available for the analysis of experimental data. Inoue and Toyozawa[2] have begun this task by giving the angular dependence of two-photon transitions in which either the initial or final state transforms according to the totally symmetric representation of the point group. Application of their results have been made by several workers.[3–5]

The present paper extends this work in two respects. First, allowed transitions between states belonging to all irreducible representations of the point group are considered. Second, the double-valued representations encountered when spin-orbit coupling is included are treated. Our attention is restricted to the center of the zone for band-to-band or exciton transitions. The results are, of course, also directly applicable to the study of point defects. Section II provides a review of the formalism of Inoue and Toyozawa and presents the proof of a theorem which permits the determination of the polarization dependence of allowed transitions between pairs of states of any symmetry from a knowledge of the polarization dependence of the transition connecting each of them to the totally symmetric state. Section III is devoted to a tabular presentation of results of the calculation. Appendix A is devoted to some symmetry properties of Clebsch-Gordan coefficients necessary to the proof of the theorem in Sec. II. Appendix B is a "dictionary" giving the translation of the labels for irreducible representations used here and in Ref. 2, into the notation used by Koster *et al.*[6]

II. FORMALISM

We consider two beams incident on the crystal, one of energy $\hbar\omega_1$ with polarization $\hat{\epsilon}_1=(l_1,m_1,n_1)$ and the other of energy $\hbar\omega_2$ with polarization $\hat{\epsilon}_2=(l_2,m_2,n_2)$. The dependence of the absorption coefficient on $\hat{\epsilon}_1$ and $\hat{\epsilon}_2$ is calculated for the 32 crystal point groups. The two-photon absorption coefficient is proportional to

$$\left|\sum_i \left\{\frac{\langle c_0|\hat{\epsilon}_1\cdot\mathbf{p}|i\rangle\langle i|\hat{\epsilon}_2\cdot\mathbf{p}|v_0\rangle}{E_i-E_{v_0}-\hbar\omega_2} + \frac{\langle c_0|\hat{\epsilon}_2\cdot\mathbf{p}|i\rangle\langle i|\hat{\epsilon}_1\cdot\mathbf{p}|v_0\rangle}{E_i-E_{v_0}-\hbar\omega_1}\right\}\right|^2. \quad (1)$$

For band-to-band transitions $|c_0\rangle$ and $|v_0\rangle$ are one-electron states in the conduction band and valence band, respectively, both at $k=0$. For a point defect they are simply the final and initial states, respectively.

Following Inoue and Toyozawa[2] we define the quantities

$$\Lambda(\omega_\alpha)=\sum_i \frac{|i\rangle\langle i|}{E_i-E_{v_0}-\hbar\omega_\alpha},$$

$$\Lambda_+=\Lambda(\omega_2)+\Lambda(\omega_1),$$

$$\Lambda_-=\Lambda(\omega_2)-\Lambda(\omega_1).$$

Equation (1) then becomes

$$|\langle c_0|[\hat{\epsilon}_1\cdot\mathbf{p}\Lambda(\omega_2)\mathbf{p}\cdot\hat{\epsilon}_2+\hat{\epsilon}_2\cdot\mathbf{p}\Lambda(\omega_1)\mathbf{p}\cdot\hat{\epsilon}_1]|v_0\rangle|^2.$$

* Research supported in part by a grant from the National Science Foundation.

[1] J. J. Hopfield and J. M. Worlock, Phys. Rev. 137, A1455 (1965).

[2] M. Inoue and Y. Toyozawa, J. Phys. Soc. Japan 20, 363 (1965).

[3] D. Fröhlich and B. Staginnus, Phys. Rev. Letters 19, 496 (1967).

[4] D. Fröhlich, B. Staginnus, and E. Schönherr, Phys. Rev. Letters 19, 1032 (1967).

[5] M. Matsuoka, J. Phys. Soc. Japan (to be published).

[6] G. F. Koster, J. O. Dimmock, R. G. Wheeler, and H. Statz, *Properties of the Thirty-Two Point Groups* (M.I.T. Press, Cambridge, Mass., 1963).

The operator in square brackets can be rewritten

$$\tfrac{1}{2}\hat{\epsilon}_1\cdot\mathbf{p}(\Lambda_+ + \Lambda_-)\mathbf{p}\cdot\hat{\epsilon}_2 + \tfrac{1}{2}\hat{\epsilon}_2\cdot\mathbf{p}(\Lambda_+ - \Lambda_-)\mathbf{p}\cdot\hat{\epsilon}_1$$
$$= \tfrac{1}{2}\{\hat{\epsilon}_1\cdot\mathbf{p}\Lambda_+\mathbf{p}\cdot\hat{\epsilon}_2 + \hat{\epsilon}_2\cdot\mathbf{p}\Lambda_+\mathbf{p}\cdot\hat{\epsilon}_1\} + \tfrac{1}{2}\{\hat{\epsilon}_1\cdot\mathbf{p}\Lambda_-\mathbf{p}\cdot\hat{\epsilon}_2 - \hat{\epsilon}_2\cdot\mathbf{p}\Lambda_-\mathbf{p}\cdot\hat{\epsilon}_1\}$$
$$= \hat{\epsilon}_1\cdot\{(\mathbf{p}\Lambda_+\mathbf{p})_{\mathrm{S}} + (\mathbf{p}\Lambda_-\mathbf{p})_{\mathrm{AS}}\}\cdot\hat{\epsilon}_2,$$

where S and AS mean the symmetric and antisymmetric parts. The tensor operator in braces, which will be labeled $\mathbf{T}$, can be written in matrix form as

$$\mathbf{T} = \begin{bmatrix} A_{xx} & A_{xy} & A_{xz} \\ A_{xy} & A_{yy} & A_{yz} \\ A_{xz} & A_{yz} & A_{zz} \end{bmatrix} + \begin{bmatrix} 0 & B_z & -B_y \\ -B_z & 0 & B_x \\ B_y & -B_x & 0 \end{bmatrix}, \quad (2)$$

where

$$B_x = \tfrac{1}{2}(p_y\Lambda_- p_z - p_z\Lambda_- p_y),$$
$$B_y = \tfrac{1}{2}(p_z\Lambda_- p_x - p_x\Lambda_- p_z),$$

and

$$B_z = \tfrac{1}{2}(p_x\Lambda_- p_y - p_y\Lambda_- p_x)$$

transform as components of a pseudovector. Also, the

$$A_{ij} = \tfrac{1}{2}(p_i\Lambda_+ p_j + p_j\Lambda_+ p_i)$$

transform as the products ij (e.g., A_{xy} transforms like xy).

We can now write expression (1) as

$$|\langle c_0|\,\hat{\epsilon}_1\cdot\mathbf{T}\cdot\hat{\epsilon}_2\,|v_0\rangle|^2,$$

where

$$\hat{\epsilon}_1\cdot\mathbf{T}\cdot\hat{\epsilon}_2 = l_1l_2A_{xx} + m_1m_2A_{yy} + n_1n_2A_{zz} + (l_1m_2 + l_2m_1)A_{xy} + (n_1l_2 + n_2l_1)A_{xz} + (m_1n_2 + m_2n_1)A_{yz} + (l_1m_2 - l_2m_1)B_z + (n_1l_2 - n_2l_1)B_y + (m_1n_2 - m_2n_1)B_x.$$

We now rewrite $\hat{\epsilon}_1\cdot\mathbf{T}\cdot\hat{\epsilon}_2$ as a linear combination of operators T_n^μ which transform under operations R of the group according to rows n of representations Γ^μ. For a transition $\Gamma^\lambda \to \Gamma^\nu$ we must calculate

$$\sum_{m,l}|\langle\lambda m|\,\hat{\epsilon}_1\cdot\mathbf{T}\cdot\hat{\epsilon}_2\,|\nu l\rangle|^2 = \sum_{m,l}|\langle\lambda m|\sum_{\mu,n,i} a_{ni}^\mu T_{ni}^\mu|\nu l\rangle|^2,$$

where $|\lambda m\rangle$ is a state transforming according to row m of Γ^λ, and similarly with $|\nu l\rangle$. The index i applies if there is more than one term transforming according to row n of Γ^μ in the decomposition of $\mathbf{T}$.

We can now state a theorem. Assume that an operator T can be decomposed into a sum of terms T_{ni}^μ which transform according to rows n and representations Γ^μ of a group, i.e., $T = \sum_{\mu,n,i} a_{ni}^\mu T_{ni}^\mu$. Then

$$\sum_{m,l}|\sum_{\mu,n,i} a_{ni}^\mu\langle\lambda m|T_{ni}^\mu|\nu l\rangle|^2 = \sum_{\mu,\tau_\mu}\gamma(\mu,\tau_\mu), \quad (3)$$

where the sum over μ on the right extends only over those representations contained in the direct product $\Gamma^{\lambda*}\times\Gamma^\nu$, and

$$\gamma(\mu,\tau_\mu) \equiv \sum_s|\langle\Gamma^1|\sum_{\sigma,n,i} a_{ni}^\sigma T_{ni}^\sigma|\mu\tau_\mu s\rangle|^2. \quad (4)$$

In Eq. (4), Γ^1 indicates a function transforming according to the identity representation and τ_μ distinguishes occurrences of representation μ, if Γ^μ is contained more than once in $\Gamma^{\lambda*}\times\Gamma^\nu$. In our context, Eq. (3) means that the angular function describing a $\Gamma^\lambda \to \Gamma^\nu$ transition is a linear combination of angular functions of the type characterizing $\Gamma^1 \to \Gamma^\mu$ transitions such that Γ^μ is contained in the decomposition of $\Gamma^{\lambda*}\times\Gamma^\nu$, and that as many distinct functions of the form for $\Gamma^1 \to \Gamma^\mu$ will occur as the number of times Γ^μ is contained in $\Gamma^{\lambda*}\times\Gamma^\nu$.

To prove the theorem we first write

$$\gamma(\mu,\tau_\mu) = \sum_s|\sum_{\sigma,n,i} a_{ni}^\sigma\langle\Gamma^1|\sum_{\lambda,\tau_\lambda,r}(\sigma n,\mu s|\lambda\tau_\lambda r)^*|\lambda\tau_\lambda r;\,\tau_\mu i\rangle|^2$$
$$= \sum_s|\sum_i a_{si}^\mu\xi_i^{\mu\tau_\mu}|^2,$$

where $\xi_i^{\mu\tau_\mu}$ are complex constants. The $(\sigma n,\mu s|\lambda\tau_\lambda r)$ are the Clebsch-Gordan coefficients defined by

$$\Psi_r^{\lambda\tau\lambda} = \sum_{n,s}(\sigma n,\mu s|\lambda\tau_\lambda r)\psi_n^\sigma\phi_s^\mu.$$

We now write

$$\sum_{m,l}|\sum_{\mu,n,i} a_{ni}^\mu\langle\lambda m|T_{ni}^\mu|\nu l\rangle|^2$$
$$= \sum_{m,l}|\sum_{\mu,n,i} a_{ni}^\mu\sum_{\sigma,\tau_\sigma,s}(\mu n,\nu l|\sigma\tau_\sigma s)^*\langle\lambda m|\sigma\tau_\sigma si\rangle|^2$$
$$= \sum_{m,l}|\sum_{\mu,n,i} a_{ni}^\mu\sum_{\tau\lambda}(\mu n,\nu l|\lambda\tau_\lambda m)^*\eta_i^{\tau\lambda}|^2,$$

where $\eta_i^{\tau\lambda}$ is a complex constant. We now make use of the following symmetry relation for Clebsch-Gordan coefficients (see Appendix A):

$$(\mu n,\nu l|\lambda\tau_\lambda m) = \sum_{\tau_\mu} C[(\lambda\mu)\bar{\nu};\,\tau_\lambda\tau_\mu](\lambda m,\bar{\nu}l|\mu\tau_\mu n)^*.$$

The $C[(\lambda\mu)\bar{\nu};\,\tau_\lambda\tau_\mu]$ are complex constants, and $\bar{\nu}$ denotes the representation which is the complex conjugate of representation ν.

We now have

$$\sum_{m,l}|\sum_{\mu,n,i} a_{ni}^\mu\sum_{\tau\lambda\tau_\mu} C[(\lambda\mu)\bar{\nu};\,\tau_\lambda\tau_\mu]^*(\lambda m,\bar{\nu}l|\mu\tau_\mu n)\eta_i^{\tau\lambda}|^2$$
$$= \sum_{m,l}\sum_{\mu,n,i}\sum_{\mu',n',i'}\sum_{\tau\lambda,\tau_\mu}\sum_{\tau\lambda'\tau_{\mu'}}(a_{ni}^\mu)(a_{n'i'}^{\mu'})^*$$
$$\times C[(\lambda\mu)\bar{\nu};\,\tau_\lambda\tau_\mu]^* C[(\lambda\mu')\bar{\nu};\,\tau_\lambda'\tau_{\mu'}'](\eta_i^{\tau\lambda})(\eta_{i'}^{\tau\lambda'})^*$$
$$\times(\lambda m,\bar{\nu}l|\mu\tau_\mu n)(\lambda m,\bar{\nu}l|\mu'\tau_{\mu'}'n')^*.$$

Performing the sum over m and l and using the orthogonality relation for the Clebsch-Gordan coefficients (see Appendix A) we find

$$\sum_{m,l}|\sum_{\mu,n,i} a_{ni}^\mu\langle\lambda m|T_{ni}^\mu|\nu l\rangle|^2$$
$$= \sum_{\mu,\tau_\mu,n}|\sum_i a_{ni}^\mu\sum_{\tau\lambda} C[(\lambda\mu)\bar{\nu};\,\tau_\lambda\tau_\mu]^*\eta_i^{\tau\lambda}|^2$$
$$= \sum_{\mu,\tau_\mu,n}|\sum_i a_{ni}^\mu\xi_i^{\mu\tau_\mu}|^2$$
$$= \sum_{\mu,\tau_\mu}\gamma(\mu,\tau_\mu),$$

as stated.

TABLE I. Angular-dependence functions. The group(s) are given at the heads of the left-hand columns. Other symbols are defined in the text.

C_1	S_2		
$A \leftrightarrow A$	$A_\pm \leftrightarrow A_\pm$		$[l_1l_2+\lambda_1m_1m_2+\lambda_2n_1n_2+\lambda_3(l_1m_2+l_2m_1)+\lambda_4(n_1l_2+n_2l_1)+\lambda_5(m_1n_2+m_2n_1)+\lambda_6(l_1m_2-l_2m_1)+\lambda_7(n_1l_2-n_2l_1)+\lambda_8(m_1n_2-m_2n_1)]^2$
$\Gamma^2 \leftrightarrow \Gamma^2$	$\Gamma_\pm{}^2 \leftrightarrow \Gamma_\pm{}^2$		
C_2	C_{1h}	C_{2h}	
$A \leftrightarrow A$	$A' \leftrightarrow A'$	$A_\pm \leftrightarrow A_\pm$	$[l_1l_2+\lambda_1m_1m_2+\lambda_2n_1n_2+\lambda_3(l_1m_2+l_2m_1)+\lambda_4(l_1m_2-l_2m_1)]^2 \equiv A(\hat{e}_1\hat{e}_2)$
$B \leftrightarrow B$	$A'' \leftrightarrow A''$	$B_\pm \leftrightarrow B_\pm$	
$A \leftrightarrow B$	$A' \leftrightarrow A''$	$A_\pm \leftrightarrow B_\pm$	$[(m_1n_2-m_2n_1)+\lambda_5(n_1l_2-n_2l_1)+\lambda_6(n_1l_2+n_2l_1)+\lambda_7(m_1n_2+m_2n_1)]^2 \equiv B(\hat{e}_1\hat{e}_2)$
$(\Gamma^3\Gamma^4) \leftrightarrow (\Gamma^3\Gamma^4)$	$(\Gamma^3\Gamma^4) \leftrightarrow (\Gamma^3\Gamma^4)$	$(\Gamma_\pm{}^3\Gamma_\pm{}^4) \leftrightarrow (\Gamma_\pm{}^3\Gamma_\pm{}^4)$	$A(\hat{e}_1\hat{e}_2)+cB(\hat{e}_1\hat{e}_2)$
D_2	C_{2v}	D_{2h}	
$A \leftrightarrow A$	$A_1 \leftrightarrow A_1$	$A_\pm \leftrightarrow A_\pm$	$[l_1l_2+\lambda_1m_1m_2+\lambda_2n_1n_2]^2 \equiv A(\hat{e}_1\hat{e}_2)$
$B_1 \leftrightarrow B_1$	$A_2 \leftrightarrow A_2$	$B_{1\pm} \leftrightarrow B_{1\pm}$	
$B_2 \leftrightarrow B_2$	$B_1 \leftrightarrow B_1$	$B_{2\pm} \leftrightarrow B_{2\pm}$	
$B_3 \leftrightarrow B_3$	$B_2 \leftrightarrow B_2$	$B_{3\pm} \leftrightarrow B_{3\pm}$	
$A \leftrightarrow B_1$	$A_1 \leftrightarrow A_2$	$A_\pm \leftrightarrow B_{1\pm}$	$[(l_1m_2+l_2m_1)+\lambda_3(l_1m_2-l_2m_1)]^2 \equiv B_1(\hat{e}_1\hat{e}_2)$
$B_2 \leftrightarrow B_3$	$B_1 \leftrightarrow B_2$	$B_{2\pm} \leftrightarrow B_{3\pm}$	
$A \leftrightarrow B_2$	$A_1 \leftrightarrow B_1$	$A_\pm \leftrightarrow B_{2\pm}$	$[(n_1l_2+n_2l_1)+\lambda_4(n_1l_2-n_2l_1)]^2 \equiv B_2(\hat{e}_1\hat{e}_2)$
$B_1 \leftrightarrow B_3$	$A_2 \leftrightarrow B_2$	$B_{1\pm} \leftrightarrow B_{3\pm}$	
$A \leftrightarrow B_3$	$A_1 \leftrightarrow B_2$	$A_\pm \leftrightarrow B_{3\pm}$	$[(m_1n_2+m_2n_1)+\lambda_5(m_1n_2-m_2n_1)]^2 \equiv B_3(\hat{e}_1\hat{e}_2)$
$B_1 \leftrightarrow B_2$	$A_2 \leftrightarrow B_1$	$B_{1\pm} \leftrightarrow B_{2\pm}$	
$\Gamma^5 \leftrightarrow \Gamma^5$	$\Gamma^5 \leftrightarrow \Gamma^5$	$\Gamma_\pm{}^5 \leftrightarrow \Gamma_\pm{}^5$	$A(\hat{e}_1\hat{e}_2)+c_1B_1(\hat{e}_1\hat{e}_2)+c_2B_2(\hat{e}_1\hat{e}_2)+c_3B_3(\hat{e}_1\hat{e}_2)$
C_4, S_4	C_{4h}		
$A \leftrightarrow A$	$A_\pm \leftrightarrow A_\pm$		$[(l_1l_2+m_1m_2)+\lambda_1n_1n_2+\lambda_2(l_1m_2-l_2m_1)]^2 \equiv A(\hat{e}_1\hat{e}_2)$
$B \leftrightarrow B$	$B_\pm \leftrightarrow B_\pm$		
$A \leftrightarrow B$	$A_\pm \leftrightarrow B_\pm$		$[(l_1l_2-m_1m_2)+\lambda_3(l_1m_2+l_2m_1)]^2 \equiv B(\hat{e}_1\hat{e}_2)$
$A \leftrightarrow E$	$A_\pm \leftrightarrow E_\pm$		$[(n_1l_2+n_2l_1)+\lambda_4(m_1n_2-m_2n_1)+\lambda_5(n_1l_2-n_2l_1)]^2+[(m_1n_2+m_2n_1)+\lambda_4(n_1l_2-n_2l_1)-\lambda_5(m_1n_2-m_2n_1)]^2 \equiv E(\hat{e}_1\hat{e}_2)$
$B \leftrightarrow E$	$B_\pm \leftrightarrow E_\pm$		
$E \leftrightarrow E$	$E_\pm \leftrightarrow E_\pm$		$A(\hat{e}_1\hat{e}_2)+cB(\hat{e}_1\hat{e}_2)$
$(\Gamma^5\Gamma^6) \leftrightarrow (\Gamma^5\Gamma^6)$	$(\Gamma_\pm{}^5\Gamma_\pm{}^6) \leftrightarrow (\Gamma_\pm{}^5\Gamma_\pm{}^6)$		$A(\hat{e}_1\hat{e}_2)+cE(\hat{e}_1\hat{e}_2)$
$(\Gamma^7\Gamma^8) \leftrightarrow (\Gamma^7\Gamma^8)$	$(\Gamma_\pm{}^7\Gamma_\pm{}^8) \leftrightarrow (\Gamma_\pm{}^7\Gamma_\pm{}^8)$		
$(\Gamma^5\Gamma^6) \leftrightarrow (\Gamma^7\Gamma^8)$	$(\Gamma_\pm{}^5\Gamma_\pm{}^6) \leftrightarrow (\Gamma_\pm{}^7\Gamma_\pm{}^8)$		$B(\hat{e}_1\hat{e}_2)+cE(\hat{e}_1\hat{e}_2)$
D_4, C_{4d}, D_{2d}	D_{4h}		
$A_1 \leftrightarrow A_1$	$A_{1\pm} \leftrightarrow A_{1\pm}$		$[(l_1l_2+m_1m_2)+\lambda_1n_1n_2]^2 \equiv A_1(\hat{e}_1\hat{e}_2)$
$A_2 \leftrightarrow A_2$	$A_{2\pm} \leftrightarrow A_{2\pm}$		
$B_1 \leftrightarrow B_1$	$B_{1\pm} \leftrightarrow B_{1\pm}$		
$B_2 \leftrightarrow B_2$	$B_{2\pm} \leftrightarrow B_{2\pm}$		
$A_1 \leftrightarrow A_2$	$A_{1\pm} \leftrightarrow A_{2\pm}$		$(l_1m_2-l_2m_1)^2 \equiv A_2(\hat{e}_1\hat{e}_2)$
$B_1 \leftrightarrow B_2$	$B_{1\pm} \leftrightarrow B_{2\pm}$		
$A_1 \leftrightarrow B_1$	$A_{1\pm} \leftrightarrow B_{1\pm}$		$(l_1l_2-m_1m_2)^2 \equiv B_1(\hat{e}_1\hat{e}_2)$
$A_2 \leftrightarrow B_2$	$A_{2\pm} \leftrightarrow B_{2\pm}$		
$A_1 \leftrightarrow B_2$	$A_{1\pm} \leftrightarrow B_{2\pm}$		$(l_1m_2+l_2m_1)^2 \equiv B_2(\hat{e}_1\hat{e}_2)$
$A_2 \leftrightarrow B_1$	$A_{2\pm} \leftrightarrow B_{1\pm}$		
$A_1 \leftrightarrow E$	$A_{1\pm} \leftrightarrow E_\pm$		$[(m_1n_2+m_2n_1)+\lambda_2(m_1n_2-m_2n_1)]^2+[(n_1l_2+n_2l_1)-\lambda_2(n_1l_2-n_2l_1)]^2 \equiv E(\hat{e}_1\hat{e}_2)$
$A_2 \leftrightarrow E$	$A_{2\pm} \leftrightarrow E_\pm$		
$B_1 \leftrightarrow E$	$B_{1\pm} \leftrightarrow E_\pm$		
$B_2 \leftrightarrow E$	$B_{2\pm} \leftrightarrow E_\pm$		
$E \leftrightarrow E$	$E_\pm \leftrightarrow E_\pm$		$A_1(\hat{e}_1\hat{e}_2)+c_1A_2(\hat{e}_1\hat{e}_2)+c_2B_1(\hat{e}_1\hat{e}_2)+c_3B_2(\hat{e}_1\hat{e}_2)$
$\Gamma^6 \leftrightarrow \Gamma^6$	$\Gamma_\pm{}^6 \leftrightarrow \Gamma_\pm{}^6$		$A_1(\hat{e}_1\hat{e}_2)+c_1A_2(\hat{e}_1\hat{e}_2)+c_2E(\hat{e}_1\hat{e}_2)$
$\Gamma^7 \leftrightarrow \Gamma^7$	$\Gamma_\pm{}^7 \leftrightarrow \Gamma_\pm{}^7$		
$\Gamma^6 \leftrightarrow \Gamma^7$	$\Gamma_\pm{}^6 \leftrightarrow \Gamma_\pm{}^7$		$B_1(\hat{e}_1\hat{e}_2)+c_1B_2(\hat{e}_1\hat{e}_2)+c_2E(\hat{e}_1\hat{e}_2)$
C_3	S_6		
$A \leftrightarrow A$	$A_\pm \leftrightarrow A_\pm$		$[(l_1l_2+m_1m_2)+\lambda_1n_1n_2+\lambda_2(l_1m_2-l_2m_1)]^2 \equiv A(\hat{e}_1\hat{e}_2)$
$\Gamma^6 \leftrightarrow \Gamma^6$	$\Gamma_\pm{}^6 \leftrightarrow \Gamma_\pm{}^6$		

TABLE I (*continued*).

C_3	S_6	
$A \leftrightarrow E$ $(\Gamma^4\Gamma^5) \leftrightarrow \Gamma^6$	$A_\pm \leftrightarrow E_\pm$ $(\Gamma_\pm^4\Gamma_\pm^5) \leftrightarrow \Gamma_\pm^6$	$[(n_1l_2+n_2l_1)+\lambda_3(l_1m_2+l_2m_1)+\lambda_4(l_1l_2-m_1m_2)+\lambda_5(m_1n_2-m_2n_1)+\lambda_6(n_1l_2-n_2l_1)]^2$ $+[(m_1n_2+m_2n_1)+\lambda_3(l_1l_2-m_1m_2)-\lambda_4(l_1m_2+l_2m_1)+\lambda_5(n_1l_2-n_2l_1)$ $-\lambda_6(m_1n_2-m_2n_1)]^2\equiv E(\hat{\epsilon}_1,\hat{\epsilon}_2)$
$E \leftrightarrow E$ $(\Gamma^4\Gamma^5) \leftrightarrow (\Gamma^4\Gamma^5)$	$E_\pm \leftrightarrow E_\pm$ $(\Gamma_\pm^4\Gamma_\pm^5) \leftrightarrow (\Gamma_\pm^4\Gamma_\pm^5)$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)+cE(\hat{\epsilon}_1\hat{\epsilon}_2)$

D_3, C_{3v}	D_{3d}	
$A_1 \leftrightarrow A_1$ $A_2 \leftrightarrow A_2$	$A_{1\pm} \leftrightarrow A_{1\pm}$ $A_{2\pm} \leftrightarrow A_{2\pm}$	$[(l_1l_2+m_1m_2)+\lambda_1n_1n_2]^2\equiv A_1(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_1 \leftrightarrow A_2$	$A_{1\pm} \leftrightarrow A_{2\pm}$	$(l_1m_2-l_2m_1)^2$
$A_1 \leftrightarrow E$ $A_2 \leftrightarrow E$ $\Gamma^4 \leftrightarrow (\Gamma^5\Gamma^6)$	$A_{1\pm} \leftrightarrow E_\pm$ $A_{2\pm} \leftrightarrow E_\pm$ $\Gamma_\pm^4 \leftrightarrow (\Gamma_\pm^5\Gamma_\pm^6)$	$[(m_1n_2-m_2n_1)+\lambda_2(m_1n_2+m_2n_1)+\lambda_3(l_1l_2-m_1m_2)]^2+[(n_1l_2-n_2l_1)-\lambda_2(n_1l_2+n_2l_1)$ $-\lambda_3(l_1m_2+l_2m_1)]^2\equiv E(\hat{\epsilon}_1\hat{\epsilon}_2)$
$E \leftrightarrow E$ $\Gamma^4 \leftrightarrow \Gamma^4$	$E_\pm \leftrightarrow E_\pm$ $\Gamma_\pm^4 \leftrightarrow \Gamma_\pm^4$	$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1A_2(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2E(\hat{\epsilon}_1\hat{\epsilon}_2)$
$(\Gamma^5\Gamma^6) \leftrightarrow (\Gamma^5\Gamma^6)$	$(\Gamma_\pm^5\Gamma_\pm^6) \leftrightarrow (\Gamma_\pm^5\Gamma_\pm^6)$	$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1A_2(\hat{\epsilon}_1\hat{\epsilon}_2)$

C_6	C_{3h}	C_{6h}	
$A \leftrightarrow A$ $B \leftrightarrow B$	$A' \leftrightarrow A'$ $A'' \leftrightarrow A''$	$A_\pm \leftrightarrow A_\pm$ $B_\pm \leftrightarrow B_\pm$	$[(l_1l_2+m_1m_2)+\lambda_1n_1n_2+\lambda_2(l_1m_2-l_2m_1)]^2\equiv A(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A \leftrightarrow B$	$A' \leftrightarrow A''$	$A_\pm \leftrightarrow B_\pm$	forbidden
$A \leftrightarrow E'$ $B \leftrightarrow E''$ $E' \leftrightarrow E''$	$A' \leftrightarrow E''$ $A'' \leftrightarrow E'$ $E' \leftrightarrow E''$	$A_\pm \leftrightarrow E_\pm'$ $B_\pm \leftrightarrow E_\pm''$ $E_\pm' \leftrightarrow E_\pm''$	$[(n_1l_2+n_2l_1)+\lambda_3(m_1n_2-m_2n_1)+\lambda_4(n_1l_2-n_2l_1)]^2$ $+[(m_1n_2+m_2n_1)+\lambda_3(n_1l_2-n_2l_1)$ $-\lambda_4(m_1n_2-m_2n_1)]^2\equiv E'(\hat{\epsilon}_1,\hat{\epsilon}_2)$
$A \leftrightarrow E''$ $B \leftrightarrow E'$	$A' \leftrightarrow E'$ $A'' \leftrightarrow E''$	$A_\pm \leftrightarrow E_\pm''$ $B_\pm \leftrightarrow E_\pm'$	$(1-n_1^2)(1-n_2^2)\equiv E''(\hat{\epsilon}_1\hat{\epsilon}_2)$
$E' \leftrightarrow E'$ $E'' \leftrightarrow E''$	$E' \leftrightarrow E'$ $E'' \leftrightarrow E''$	$E_\pm' \leftrightarrow E_\pm'$ $E_\pm'' \leftrightarrow E_\pm''$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)+cE''(\hat{\epsilon}_1\hat{\epsilon}_2)$
$(\Gamma^7\Gamma^8) \leftrightarrow (\Gamma^7\Gamma^8)$ $(\Gamma^9\Gamma^{10}) \leftrightarrow (\Gamma^9\Gamma^{10})$	$(\Gamma^7\Gamma^8) \leftrightarrow (\Gamma^7\Gamma^8)$ $(\Gamma^9\Gamma^{10}) \leftrightarrow (\Gamma^9\Gamma^{10})$	$(\Gamma_\pm^7\Gamma_\pm^8) \leftrightarrow (\Gamma_\pm^7\Gamma_\pm^8)$ $(\Gamma_\pm^9\Gamma_\pm^{10}) \leftrightarrow (\Gamma_\pm^9\Gamma_\pm^{10})$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)+cE'(\hat{\epsilon}_1\hat{\epsilon}_2)$
$(\Gamma^7\Gamma^8) \leftrightarrow (\Gamma^9\Gamma^{10})$	$(\Gamma^7\Gamma^8) \leftrightarrow (\Gamma^9\Gamma^{10})$	$(\Gamma_\pm^7\Gamma_\pm^8) \leftrightarrow (\Gamma_\pm^9\Gamma_\pm^{10})$	$E''(\hat{\epsilon}_1\hat{\epsilon}_2)$
$(\Gamma^7\Gamma^8) \leftrightarrow (\Gamma^{11}\Gamma^{12})$ $(\Gamma^9\Gamma^{10}) \leftrightarrow (\Gamma^{11}\Gamma^{12})$	$(\Gamma^7\Gamma^8) \leftrightarrow (\Gamma^{11}\Gamma^{12})$ $(\Gamma^9\Gamma^{10}) \leftrightarrow (\Gamma^{11}\Gamma^{12})$	$(\Gamma_\pm^7\Gamma_\pm^8) \leftrightarrow (\Gamma_\pm^{11}\Gamma_\pm^{12})$ $(\Gamma_\pm^9\Gamma_\pm^{10}) \leftrightarrow (\Gamma_\pm^{11}\Gamma_\pm^{12})$	$E'(\hat{\epsilon}_1\hat{\epsilon}_2)+cE''(\hat{\epsilon}_1\hat{\epsilon}_2)$
$(\Gamma^{11}\Gamma^{12}) \leftrightarrow (\Gamma^{11}\Gamma^{12})$	$(\Gamma^{11}\Gamma^{12}) \leftrightarrow (\Gamma^{11}\Gamma^{12})$	$(\Gamma_\pm^{11}\Gamma_\pm^{12}) \leftrightarrow (\Gamma_\pm^{11}\Gamma_\pm^{12})$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)$

D_6, C_{6v}	D_{3h}	D_{6h}	
$A_1 \leftrightarrow A_1$ $A_2 \leftrightarrow A_2$ $B_1 \leftrightarrow B_1$ $B_2 \leftrightarrow B_2$	$A_1' \leftrightarrow A_1'$ $A_2' \leftrightarrow A_2'$ $A_1'' \leftrightarrow A_1''$ $A_2'' \leftrightarrow A_2''$	$A_{1\pm} \leftrightarrow A_{1\pm}$ $A_{2\pm} \leftrightarrow A_{2\pm}$ $B_{1\pm} \leftrightarrow B_{1\pm}$ $B_{2\pm} \leftrightarrow B_{2\pm}$	$[(l_1l_2+m_1m_2)+\lambda_1n_1n_2]^2\equiv A_1(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_1 \leftrightarrow A_2$ $B_1 \leftrightarrow B_2$	$A_1' \leftrightarrow A_2'$ $A_1'' \leftrightarrow A_2''$	$A_{1\pm} \leftrightarrow A_{2\pm}$ $B_{1\pm} \leftrightarrow B_{2\pm}$	$(l_1m_2-l_2m_1)^2\equiv A_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_1 \leftrightarrow B_1$ $A_1 \leftrightarrow B_2$ $A_2 \leftrightarrow B_1$ $A_2 \leftrightarrow B_2$	$A_1' \leftrightarrow A_1''$ $A_1' \leftrightarrow A_2''$ $A_2' \leftrightarrow A_1''$ $A_2' \leftrightarrow A_2''$	$A_{1\pm} \leftrightarrow B_{1\pm}$ $A_{1\pm} \leftrightarrow B_{2\pm}$ $A_{2\pm} \leftrightarrow B_{1\pm}$ $A_{2\pm} \leftrightarrow B_{2\pm}$	forbidden
$A_1 \leftrightarrow E_1$ $A_2 \leftrightarrow E_1$ $B_1 \leftrightarrow E_2$ $B_2 \leftrightarrow E_2$ $E_1 \leftrightarrow E_2$	$A_1' \leftrightarrow E''$ $A_2' \leftrightarrow E''$ $A_2'' \leftrightarrow E'$ $A_1'' \leftrightarrow E'$ $E' \leftrightarrow E''$	$A_{1\pm} \leftrightarrow E_{1\pm}$ $A_{2\pm} \leftrightarrow E_{1\pm}$ $B_{1\pm} \leftrightarrow E_{2\pm}$ $B_{2\pm} \leftrightarrow E_{2\pm}$ $E_{1\pm} \leftrightarrow E_{2\pm}$	$[(m_1n_2-m_2n_1)+\lambda_2(m_1n_2+m_2n_1)]^2+[(n_1l_2-n_2l_1)-\lambda_2(n_1l_2+n_2l_1)]^2\equiv E_1(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_1 \leftrightarrow E_2$ $A_2 \leftrightarrow E_2$ $B_1 \leftrightarrow E_1$ $B_2 \leftrightarrow E_1$	$A_1' \leftrightarrow E'$ $A_2' \leftrightarrow E'$ $A_2'' \leftrightarrow E''$ $A_1'' \leftrightarrow E''$	$A_{1\pm} \leftrightarrow E_{2\pm}$ $A_{2\pm} \leftrightarrow E_{2\pm}$ $B_{1\pm} \leftrightarrow E_{1\pm}$ $B_{2\pm} \leftrightarrow E_{1\pm}$	$(1-n_1^2)(1-n_2^2)\equiv E_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$E_1 \leftrightarrow E_1$ $E_2 \leftrightarrow E_2$	$E'' \leftrightarrow E''$ $E' \leftrightarrow E'$	$E_{1\pm} \leftrightarrow E_{1\pm}$ $E_{2\pm} \leftrightarrow E_{2\pm}$	$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1A_2(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2E_2(\hat{\epsilon}_1\hat{\epsilon}_2)$

TABLE I (*continued*).

D_6, C_{6v}	D_{3h}	D_{6h}	
$\Gamma^7 \leftrightarrow \Gamma^7$	$\Gamma^7 \leftrightarrow \Gamma^7$	$\Gamma_\pm{}^7 \leftrightarrow \Gamma_\pm{}^7$	$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1A_2(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2E_1(\hat{\epsilon}_1\hat{\epsilon}_2)$
$\Gamma^8 \leftrightarrow \Gamma^8$	$\Gamma^8 \leftrightarrow \Gamma^8$	$\Gamma_\pm{}^8 \leftrightarrow \Gamma_\pm{}^8$	
$\Gamma^7 \leftrightarrow \Gamma^8$	$\Gamma^7 \leftrightarrow \Gamma^8$	$\Gamma_\pm{}^7 \leftrightarrow \Gamma_\pm{}^8$	$E_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$\Gamma^7 \leftrightarrow \Gamma^9$	$\Gamma^7 \leftrightarrow \Gamma^9$	$\Gamma_\pm{}^7 \leftrightarrow \Gamma_\pm{}^9$	$E_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1E_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$\Gamma^8 \leftrightarrow \Gamma^9$	$\Gamma^8 \leftrightarrow \Gamma^9$	$\Gamma_\pm{}^8 \leftrightarrow \Gamma_\pm{}^9$	
$\Gamma^9 \leftrightarrow \Gamma^9$	$\Gamma^9 \leftrightarrow \Gamma^9$	$\Gamma_\pm{}^9 \leftrightarrow \Gamma_\pm{}^9$	$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1A_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
T		T_h	
$A \leftrightarrow A$		$A_\pm \leftrightarrow A_\pm$	$(l_1l_2+m_1m_2+n_1n_2)^2=(\hat{\epsilon}_1\cdot\hat{\epsilon}_2)^2\equiv A(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A \leftrightarrow E$		$A_\pm \leftrightarrow E_\pm$	$(l_1{}^2l_2{}^2+m_1{}^2m_2{}^2+n_1{}^2n_2{}^2)-(l_1l_2m_1m_2+m_1m_2n_1n_2+n_1n_2l_1l_2)\equiv E(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A \leftrightarrow T$		$A_\pm \leftrightarrow T_\pm$	$[(m_1n_2-m_2n_1)+\lambda(m_1n_2+m_2n_1)]^2+[(n_1l_2-n_2l_1)+\lambda(n_1l_2+n_2l_1)]^2+[(l_1m_2-l_2m_1)$
$E \leftrightarrow T$		$E_\pm \leftrightarrow T_\pm$	$+\lambda(l_1m_2+l_2m_1)]^2\equiv T(\hat{\epsilon}_1\hat{\epsilon}_2,\lambda)$
$E \leftrightarrow E$		$E_\pm \leftrightarrow E_\pm$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)+cE(\hat{\epsilon}_1\hat{\epsilon}_2)$
$T \leftrightarrow T$		$T_\pm \leftrightarrow T_\pm$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1E(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2T(\hat{\epsilon}_1\hat{\epsilon}_2,\lambda)+c_3T(\hat{\epsilon}_1\hat{\epsilon}_2,\lambda')$
$\Gamma^5 \leftrightarrow \Gamma^5$		$\Gamma_\pm{}^5 \leftrightarrow \Gamma_\pm{}^5$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)+cT(\hat{\epsilon}_1\hat{\epsilon}_2,\lambda)$
$\Gamma^5 \leftrightarrow (\Gamma^6\Gamma^7)$		$\Gamma_\pm{}^5 \leftrightarrow (\Gamma_\pm{}^6\Gamma_\pm{}^7)$	$E(\hat{\epsilon}_1\hat{\epsilon}_2)+cT(\hat{\epsilon}_1\hat{\epsilon}_2,\lambda)$
$(\Gamma^6\Gamma^7) \leftrightarrow (\Gamma^6\Gamma^7)$		$(\Gamma_\pm{}^6\Gamma_\pm{}^7) \leftrightarrow (\Gamma_\pm{}^6\Gamma_\pm{}^7)$	$A(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1E(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2T(\hat{\epsilon}_1\hat{\epsilon}_2,\lambda)$
O, T_d	O_h		
$A_1 \leftrightarrow A_1$	$A_{1\pm} \leftrightarrow A_{1\pm}$		$(l_1l_2+m_1m_2+n_1n_2)^2=(\hat{\epsilon}_1\cdot\hat{\epsilon}_2)^2\equiv A_1(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_2 \leftrightarrow A_2$	$A_{2\pm} \leftrightarrow A_{2\pm}$		
$A_1 \leftrightarrow A_2$	$A_{1\pm} \leftrightarrow A_{2\pm}$		forbidden
$A_1 \leftrightarrow E$	$A_{1\pm} \leftrightarrow E_\pm$		$(l_1{}^2l_2{}^2+m_1{}^2m_2{}^2+n_1{}^2n_2{}^2)-(l_1l_2m_1m_2+m_1m_2n_1n_2+n_1n_2l_1l_2)\equiv E(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_2 \leftrightarrow E$	$A_{2\pm} \leftrightarrow E_\pm$		
$A_1 \leftrightarrow T_1$	$A_{1\pm} \leftrightarrow T_{1\pm}$		$(m_1m_2-m_2n_1)^2+(l_2n_1-l_1n_2)^2+(l_1m_2-l_2m_1)^2=(\hat{\epsilon}_1\times\hat{\epsilon}_2)^2\equiv T_1(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_2 \leftrightarrow T_2$	$A_{2\pm} \leftrightarrow T_{2\pm}$		
$A_1 \leftrightarrow T_2$	$A_{1\pm} \leftrightarrow T_{2\pm}$		$(l_1m_2+l_2m_1)^2+(m_1n_2+m_2n_1)^2+(l_1n_2+l_2n_1)^2\equiv T_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$A_2 \leftrightarrow T_1$	$A_{2\pm} \leftrightarrow T_{1\pm}$		
$E \leftrightarrow E$	$E_\pm \leftrightarrow E_\pm$		$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+cE(\hat{\epsilon}_1\hat{\epsilon}_2)$
$E \leftrightarrow T_1$	$E_\pm \leftrightarrow T_{1\pm}$		$T_1(\hat{\epsilon}_1\hat{\epsilon}_2)+cT_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$E \leftrightarrow T_2$	$E_\pm \leftrightarrow T_{2\pm}$		
$T_1 \leftrightarrow T_1$	$T_{1\pm} \leftrightarrow T_{1\pm}$		$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1E(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2T_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_3T_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$T_2 \leftrightarrow T_2$	$T_{2\pm} \leftrightarrow T_{2\pm}$		
$T_1 \leftrightarrow T_2$	$T_{1\pm} \leftrightarrow T_{2\pm}$		$E(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1T_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2T_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$\Gamma^6 \leftrightarrow \Gamma^6$	$\Gamma_\pm{}^6 \leftrightarrow \Gamma_\pm{}^6$		$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+cT_1(\hat{\epsilon}_1\hat{\epsilon}_2)$
$\Gamma^7 \leftrightarrow \Gamma^7$	$\Gamma_\pm{}^7 \leftrightarrow \Gamma_\pm{}^7$		
$\Gamma^6 \leftrightarrow \Gamma^7$	$\Gamma_\pm{}^6 \leftrightarrow \Gamma_\pm{}^7$		$T_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$\Gamma^6 \leftrightarrow \Gamma^8$	$\Gamma_\pm{}^6 \leftrightarrow \Gamma_\pm{}^8$		$E(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1T_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2T_2(\hat{\epsilon}_1\hat{\epsilon}_2)$
$\Gamma^7 \leftrightarrow \Gamma^8$	$\Gamma_\pm{}^7 \leftrightarrow \Gamma_\pm{}^8$		
$\Gamma^8 \leftrightarrow \Gamma^8$	$\Gamma_\pm{}^8 \leftrightarrow \Gamma_\pm{}^8$		$A_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_1E(\hat{\epsilon}_1\hat{\epsilon}_2)+c_2T_1(\hat{\epsilon}_1\hat{\epsilon}_2)+c_3T_2(\hat{\epsilon}_1\hat{\epsilon}_2)$

III. RESULTS

The angular functions for all possible band-to-band transitions are presented in Table I. There the λ_i are real constants and c_i are real positive constants. In groups having inversion symmetry, the parity cannot change in two-photon absorption so that only transitions like $\Gamma_+{}^\mu \rightarrow \Gamma_+{}^\nu$ and $\Gamma_-{}^\mu \rightarrow \Gamma_-{}^\nu$ are allowed. Two representations in parentheses indicates that they are complex conjugates of each other and are degenerate. The single-valued representations are labeled by the usual symbols for molecular applications, i.e., A, B, E, and T. The double-valued representations are labeled by Γ^μ and these are defined by Koster, Dimmock, Wheeler, and Statz.[6] Their notation for the single-valued representations is tabulated along with ours in Appendix B for convenience. The diagrams in Figs. 1–3 indicate the axes with respect to which $\hat{\epsilon}_1$ and $\hat{\epsilon}_2$ are defined.

The angular functions for transitions of the type $\Gamma^1 \leftrightarrow \Gamma^\mu$ have been recalculated and agree with those reported by Inoue and Toyozawa except for the following cases: the transitions of the type $\Gamma^1 \leftrightarrow \Gamma^E$, where E denotes the complex-conjugate pair of one-dimensional representations (the basis functions of which are degenerate by time reversal), for the groups

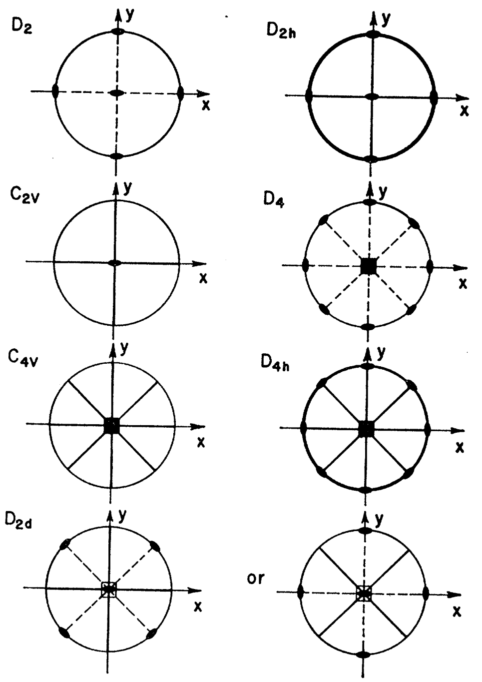

FIG. 1. Diagrams to define the axes for the point groups D_2, D_{2h}, C_{2v}, D_4, C_{4v}, D_{4h}, and D_{2d}. (For groups C_n, S_n, and C_{nh} the z axis is the axis of rotation, with x and y arbitrary.)

C_4, C_{4h}, S_4, C_3, S_6, C_6, C_{6h}, and C_{3h}. The disagreement seems to be the result of an algebraic error in the earlier work.

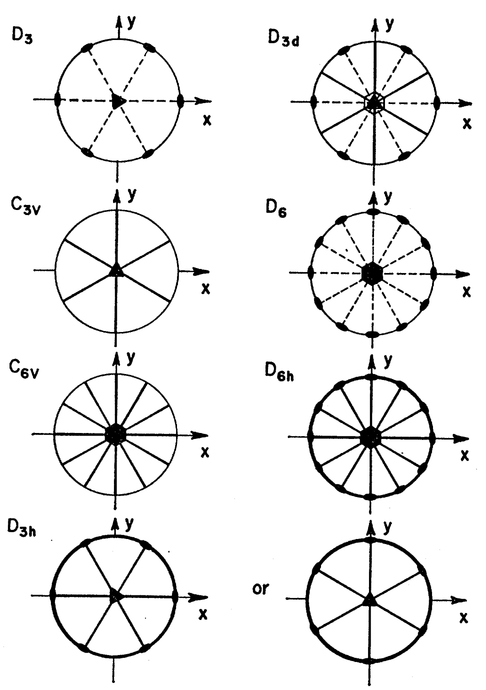

FIG. 2. Diagram to define the axes for the point groups D_3, D_{3d}, C_{3v}, D_6, C_{6v}, D_{6h}, and D_{3h}.

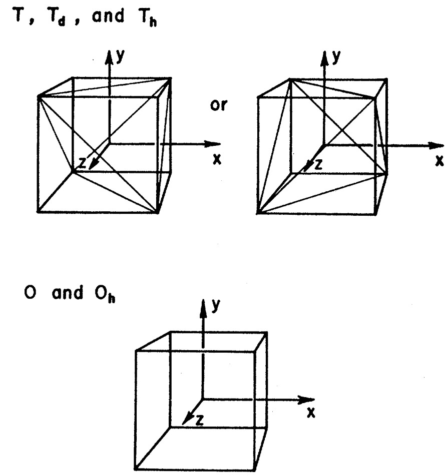

FIG. 3. Diagram to define the axes for the point groups T, T_d, T_h, O, and O_h.

ACKNOWLEDGMENTS

We wish to thank Professor M. Inoue and Professor Y. Toyozawa for a most helpful correspondence on this point.

APPENDIX A: DERIVATION OF THE CLEBSCH-GORDAN SYMMETRY RELATION

Since[7] the Clebsch-Gordan coefficients are elements of a unitary matrix, they obey the orthogonality relations

$$\sum_{j,l}(\mu j,\nu l|\lambda\tau_\lambda s)^*(\mu j,\nu l|\lambda'\tau_{\lambda'}'s')=\delta(\lambda,\lambda')\delta(\tau_\lambda,\tau_{\lambda'}')\delta(s,s')$$

and

$$\sum_{\lambda,\tau_\lambda,s}(\mu j,\nu l|\lambda\tau_\lambda s)^*(\mu j',\nu l'|\lambda\tau_\lambda s)=\delta(j,j')\delta(l,l').$$

From the definition of Clebsch-Gordan coefficients, we can write

$$\Gamma_{s'm}^{\lambda}(R)\Gamma_{kl}^{\nu}(R)=\sum_{\rho,\tau_\rho}\sum_{p,j}(\lambda m,\nu l|\rho\tau_\rho j)^*\Gamma_{pj}^{\rho\tau_\rho}(R)\times(\lambda s',\nu k|\rho\tau_\rho p)\quad\text{(A1)}$$

and

$$\Gamma_{ij}^{\mu}(R)\Gamma_{kl'}^{\nu}(R)=\sum_{\gamma,\tau_\gamma}\sum_{ss'}(\mu j,\nu l'|\gamma\tau_\gamma s)^*\Gamma_{s's}^{\gamma\tau_\gamma}(R)\times(\mu i,\nu k|\gamma\tau_\gamma s').\quad\text{(A2)}$$

Multiplying (A2) by $\Gamma_{kl}^{\nu}(R)^*$ and summing over k gives

$$\Gamma_{ij}^{\mu}(R)\delta(l,l')=\sum_{\gamma,\tau_\gamma}\sum_{s,s',k}(\mu j,\nu l'|\gamma\tau_\gamma s)^*\Gamma_{s's}^{\gamma}(R)\times\Gamma_{kl}^{\nu}(R)^*(\mu i,\nu k|\gamma\tau_\gamma s'),$$

[7] This follows the treatment given by M. Hamermesh [*Group Theory* (Addison-Wesley Publishing Co., Reading, Mass., 1962), p. 260] for real representations in his discussion of the symmetric group.

where it has been observed that $\Gamma_{s's}{}^{\gamma\tau_\gamma}(R)=\Gamma_{s's}{}^{\gamma}(R)$. Multiply this by $(\mu j,\nu l'|\lambda\tau_\lambda m)$ and sum over j and l':

$$\sum_j(\mu j,\nu l|\lambda\tau_\lambda m)\Gamma_{ij}{}^{\mu}(R)=\sum_{s'k}\Gamma_{s'm}{}^{\lambda}(R)\Gamma_{kl}{}^{\nu}(R)^*(\mu i,\nu k|\lambda\tau_\lambda s').\quad\text{(A3)}$$

Changing ν to $\bar\nu$ in (A1) and putting this into (A3), we obtain

$$\sum_j(\mu j,\nu l|\lambda\tau_\lambda m)\Gamma_{ij}{}^{\mu}(R)=\sum_{\rho,\tau_\rho}\sum_{p,j,s',k}(\lambda m,\bar\nu l|\rho\tau_\rho j)^*\Gamma_{pj}{}^{\rho}(R)\times(\lambda s',\bar\nu k|\rho\tau_\rho p)(\mu i,\nu k|\lambda\tau_\lambda s').$$

Now multiply by $(\lambda m,\bar\nu l|\sigma\tau_\sigma n)$ and sum over m and l:

$$\sum_{j,m,l}\Gamma_{ij}{}^{\mu}(R)(\mu j,\nu l|\lambda\tau_\lambda m)(\lambda m,\bar\nu l|\sigma\tau_\sigma n)=\sum_{p,s',k}(\lambda s',\bar\nu k|\sigma\tau_\sigma p)(\mu i,\nu k|\lambda\tau_\lambda s')\Gamma_{pn}{}^{\sigma}(R).$$

This can be written

$$\sum_j\Gamma_{ij}{}^{\mu}(R)M_{jn}=\sum_p M_{ip}\Gamma_{pn}{}^{\sigma}(R),$$

where

$$M_{jn}=\sum_{m,l}(\mu j,\nu l|\lambda\tau_\lambda m)(\lambda m,\bar\nu l|\sigma\tau_\sigma n),$$

or

$$\mathbf{\Gamma}^{\mu}(R)\mathbf{M}=\mathbf{M}\mathbf{\Gamma}^{\sigma}(R).$$

If $\mathbf{\Gamma}^{\mu}$ and $\mathbf{\Gamma}^{\sigma}$ are inequivalent, then Schur's lemma tells us that $\mathbf{M}$ is the null matrix. If $\mathbf{\Gamma}^{\mu}$ is equivalent to $\mathbf{\Gamma}^{\sigma}$, then $\mathbf{M}$ is a constant multiple of the unit matrix:

$$\sum_{m,l}(\mu j,\nu l|\lambda\tau_\lambda m)(\lambda m,\bar\nu l|\sigma\tau_\sigma n)=C[(\lambda\mu)\bar\nu;\,\tau_\lambda\tau_\mu]\delta(\sigma,\mu)\delta(n,j),$$

where the constant on the right depends on λ, μ, ν, τ_λ, and τ_μ, and the parentheses in the square brackets indicate the pairs of indices which are interchanged in the Clebsch-Gordan coefficients. Bringing the second factor on the left side to the right we have the result

$$(\mu n,\nu l|\lambda\tau_\lambda m)=\sum_{\tau_\mu}C[(\lambda\mu)\bar\nu;\,\tau_\lambda\tau_\mu](\lambda m,\bar\nu l|\mu\tau_\mu n)^*,$$

used in Sec. II.

APPENDIX B: DICTIONARY OF NOTATIONS

A brief dictionary of corresponding notations is given in Table II.

TABLE II. Correspondence of present notation with that of Koster *et al.*[a] for the single-valued representations.

Group	C_1	S_2	C_2		C_{1h}		C_{2h}		D_2				C_{2v}			
Present notation	A	$A_\pm$	A	B	A'	A''	$A_\pm$	$B_\pm$	A	B_1	B_2	B_3	A_1	A_2	B_1	B_2
Notation of (6)	Γ_1	$\Gamma_1^\pm$	Γ_1	Γ_2	Γ_1	Γ_2	$\Gamma_1^\pm$	$\Gamma_2^\pm$	Γ_1	Γ_3	Γ_2	Γ_4	Γ_1	Γ_3	Γ_2	Γ_4

D_{2h}				C_4			S_4			C_{4h}			D_4				
$A_\pm$	$B_{1\pm}$	$B_{2\pm}$	$B_{3\pm}$	A	B	E	A	B	E	$A_\pm$	$B_\pm$	$E_\pm$	A_1	A_2	B_1	B_2	E
$\Gamma_1^\pm$	$\Gamma_3^\pm$	$\Gamma_2^\pm$	$\Gamma_4^\pm$	Γ_1	Γ_2	$(\Gamma_3\Gamma_4)$	Γ_1	Γ_2	$(\Gamma_3\Gamma_4)$	$\Gamma_1^\pm$	$\Gamma_2^\pm$	$(\Gamma_3^\pm\Gamma_4^\pm)$	Γ_1	Γ_2	Γ_3	Γ_4	Γ_5

C_{4v}					D_{2d}					D_{4h}					C_3		S_6	
A_1	A_2	B_1	B_2	E	A_1	A_2	B_1	B_2	E	$A_{1\pm}$	$A_{2\pm}$	$B_{1\pm}$	$B_{2\pm}$	$E_\pm$	A	E	$A_\pm$	$E_\pm$
Γ_1	Γ_2	Γ_3	Γ_4	Γ_5	Γ_1	Γ_2	Γ_3	Γ_4	Γ_5	$\Gamma_1^\pm$	$\Gamma_2^\pm$	$\Gamma_3^\pm$	$\Gamma_4^\pm$	$\Gamma_5^\pm$	Γ_1	$(\Gamma_2\Gamma_3)$	$\Gamma_1^\pm$	$(\Gamma_2^\pm\Gamma_3^\pm)$

D_3			C_{3v}			D_{3d}			C_6				C_{3h}			
A_1	A_2	E	A_1	A_2	E	$A_{1\pm}$	$A_{2\pm}$	$E_\pm$	A	B	E'	E''	A'	A''	E'	E''
Γ_1	Γ_2	Γ_3	Γ_1	Γ_2	Γ_3	$\Gamma_1^\pm$	$\Gamma_2^\pm$	$\Gamma_3^\pm$	Γ_1	Γ_4	$(\Gamma_5\Gamma_6)$	$(\Gamma_2\Gamma_3)$	Γ_1	Γ_4	$(\Gamma_2\Gamma_3)$	$(\Gamma_5\Gamma_6$

C_{6h}				D_6						C_{6v}						D_{3h}					
$A_\pm$	$B_\pm$	$E_\pm'$	$E_\pm''$	A_1	A_2	B_1	B_2	E_1	E_2	A_1	A_2	B_1	B_2	E_1	E_2	A_1'	A_2'	A_1''	A_2''	E'	E''
$\Gamma_1^\pm$	$\Gamma_4^\pm$	$(\Gamma_5^\pm\Gamma_6^\pm)$	$(\Gamma_2^\pm\Gamma_3^\pm)$	Γ_1	Γ_2	Γ_3	Γ_4	Γ_5	Γ_6	Γ_1	Γ_2	Γ_4	Γ_3	Γ_5	Γ_6	Γ_1	Γ_2	Γ_3	Γ_4	Γ_6	Γ_5

D_{6h}						T			T_h			O				
$A_{1\pm}$	$A_{2\pm}$	$B_{1\pm}$	$B_{2\pm}$	$E_{1\pm}$	$E_{2\pm}$	A	E	T	$A_\pm$	$E_\pm$	$T_\pm$	A_1	A_2	E	T_1	T_2
$\Gamma_1^\pm$	$\Gamma_2^\pm$	$\Gamma_3^\pm$	$\Gamma_4^\pm$	$\Gamma_5^\pm$	$\Gamma_6^\pm$	Γ_1	$(\Gamma_2\Gamma_3)$	Γ_4	$\Gamma_1^\pm$	$(\Gamma_2^\pm\Gamma_3^\pm)$	$\Gamma_4^\pm$	Γ_1	Γ_2	Γ_3	Γ_4	Γ_5

T_d					O_h				
A_1	A_2	E	T_1	T_2	$A_{1\pm}$	$A_{2\pm}$	$E_\pm$	$T_{1\pm}$	$T_{2\pm}$
Γ_1	Γ_2	Γ_3	Γ_4	Γ_5	$\Gamma_1^\pm$	$\Gamma_2^\pm$	$\Gamma_3^\pm$	$\Gamma_4^\pm$	$\Gamma_5^\pm$

[a] See Ref. 6.