

## Polarization Dependence of Three-Photon Absorption in Solids

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ABSTRACT. The polarization dependence of three-photon absorption in solids is developed using the Wigner-Eckart theorem for finite symmetry groups. The expression for the absorption coefficient is separated into two parts: i) The geometrical part, which depends on the polarization vectors of the three photons. ii) The dynamical part, which contains the product of three electron-photon matrix elements. New information can be gained by studying these two parts. An application to TiCl crystal is given and an experimental method is suggested.

### I. Introduction

Unlike ordinary one-photon spectroscopy, multiphoton spectroscopy has the advantage that the absorption coefficient depends on the direction of the polarization vectors even in a cubic crystal. Multiphoton measurements are thus an important tool for studying electronic structure in solids since the polarization dependence of multiphoton absorption allows symmetry assignments of the electronic states involved (*see*, for instance, the review articles of Worlock 1972 and Bassani 1972, and references quoted therein).

The polarization dependence of two-photon absorption has been calculated by Inoue and Toyozawa (1965) and Bader and Gold (1968), who explicitly include the double group representations. Denisov and Makarov (1973) also include the effect of time-reversal symmetry. Polarization dependence of two-photon absorption has been observed experimentally by a number of authors (Frohlich *et al.*

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1967, Frohlich and Stagninus 1967, Frohlich *et al.* 1968, Park and Stafford 1969, Pradere and Mysyrowicz 1970, Macsuoka 1967 and Frohlich *et al.* 1970). Three-photon absorption co-efficient has been calculated by Bassani and Hassan (1972) using perturbation theory and the band model which takes into account the allowed electron-photon matrix elements. The methods of Inoue and Toyozawa for calculating the polarization dependence of two-photon transition has been extended to the case of three-photon transitions by Badrysheva and Moskalenko (1966).

In this paper, we develop a procedure to study the polarization dependence of a three-photon electronic interband transition. The procedure is based on applying the Wigner-Eckart theorem to finite symmetry groups (Koster 1958). Doni *et al.* (1974) used this method for the case of two-photon absorption. We divide the expressions for the absorption coefficient into a geometrical part, which depends on the photon polarization, and a dynamical part, which contains appropriate reduced matrix elements of the momentum operator. We show that the symmetry of the crystal determines not only the polarization dependence of a given final state, but also supplies very useful information on the strength of contribution with different symmetries. Finally we apply the procedure to the case of TlCl crystal.

## II General Definitions

From third-order time-dependent perturbation theory, one obtains the following expression (Bassani and Hassan 1972) for the absorption coefficient of photon ( $\hbar\omega_1$ ) in the presence of the photons ( $\hbar\omega_2$ ) and ( $\hbar\omega_3$ ):

$$\begin{aligned} \alpha(\omega_1) = & \sum_{f_i^u} \left| \sum_{\varphi_m^v, \psi_n^\lambda} \mathbf{M}_{f_0} \{ (1 + \mathcal{P}_{12}) \times \frac{\underline{\varepsilon}_3 \cdot \underline{\varepsilon}_1 \cdot \underline{\varepsilon}_2}{(E_{\varphi_m^v} - E_0 - \hbar\omega_1 - \hbar\omega_2)(E_{\psi_n^\lambda} - E_0 - \hbar\omega_2)} \right. \\ & + (1 + \mathcal{P}_{23}) \frac{\underline{\varepsilon}_1 \cdot \underline{\varepsilon}_2 \cdot \underline{\varepsilon}_3}{(E_{\varphi_m^v} - E_0 - \hbar\omega_2 - \hbar\omega_3)(E_{\psi_n^\lambda} - E_0 - \hbar\omega_3)} \\ & \left. + (1 + \mathcal{P}_{13}) \frac{\underline{\varepsilon}_2 \cdot \underline{\varepsilon}_3 \cdot \underline{\varepsilon}_1}{(E_{\varphi_m^v} - E_0 - \hbar\omega_1 - \hbar\omega_3)(E_{\psi_n^\lambda} - E_0 - \hbar\omega_1)} \right\}^2 \times \\ & \times \delta(E_{f_i^u} - E_0 - \hbar\omega_1 - \hbar\omega_2 - \hbar\omega_3) \end{aligned} \quad (1)$$

In the above expression we write

$$C = \frac{32 \pi e^6 \hbar^4 N_2 N_3}{\text{cm}^6 n_1 n_2^2 n_3^2 \omega_1 \omega_2 \omega_3}$$

while

$$\mathbf{M}_{f_0} = \langle f_i^u | \mathbf{P} | \varphi_m^v \rangle \langle \varphi_m^v | \mathbf{P} | \psi_n^\lambda \rangle \langle \psi_n^\lambda | \mathbf{P} | 0 \rangle$$

is the product of three electron-photon matrix elements between the ground states  $|0\rangle$  and a final state  $|f_l^\mu\rangle$  of the crystal (with energy  $E_{f_l^\mu}$ ) which belongs to the  $l^{\text{th}}$  row of the irreducible representation  $D^\mu$  of the little group at  $\underline{k} = 0$ . The sum over  $f_l^\mu$  includes all final states with any symmetry  $(\mu, l)$ .  $|\varphi_m^\nu\rangle$  and  $|\psi_n^\lambda\rangle$  indicate two different intermediate states (with energies  $E_{\varphi_m^\nu}$  and  $E_{\psi_n^\lambda}$ ) belonging respectively to the  $m^{\text{th}}$  and  $n^{\text{th}}$  row of the irreducible representation  $D^\mu$  and  $D^\lambda$  at  $k = 0$ . The sum over  $\varphi_m^\nu$  and  $\psi_n^\lambda$  runs over all intermediate states with any symmetry  $(\nu, m)$  and  $(\lambda, n)$ . The total momentum operator  $\underline{P}$  is given by  $\underline{P} = \sum_i \underline{p}_i$ , where  $\underline{p}_i$  is the momentum operator for the  $i^{\text{th}}$  electron.

$\mathcal{P}_{ij}$  interchanges  $\underline{\varepsilon}_i$  with  $\underline{\varepsilon}_j$  and  $\omega_i$  with  $\omega_j$  (the photon polarization and frequencies respectively). We note explicitly that the only representations  $D^\mu$ ,  $D^\nu$  and  $D^\lambda$  of interest are those of the single group at  $\underline{k} = 0$ , which coincide with the representations of the single point group of the crystal.

The standard way (Badrysheva and Moskalenko 1966) to handle expression (1) is to define the quantities

$$\Lambda(\omega_i) = \sum_{\psi_n^\lambda} \frac{|\psi_n^\lambda\rangle\langle\psi_n^\lambda|}{E_{\psi_n^\lambda} - E_0 - \hbar\omega_i} \quad (2a)$$

$$\Lambda_\pm = \Lambda(\omega_i) \pm \Lambda(\omega_j), \quad (2b)$$

and

$$\Lambda(\omega_{ij}) = \sum_{\varphi_m^\nu} \frac{|\varphi_m^\nu\rangle\langle\varphi_m^\nu|}{E_{\varphi_m^\nu} - E_0 - \hbar\omega_i - \hbar\omega_j}, \quad \text{with } i, j = 1, 2, 3, \quad (3)$$

containing contributions from all intermediate states and to determine their symmetry properties. Due to the summation over all the intermediate valence and conduction states, it is difficult to evaluate the strengths with which the various angular dependence functions appear in the absorption coefficient. However, the energy denominator, in the above expression, dictates that the largest contribution to the absorption rate comes from intermediate states which are very close to the conduction or valence bands. Therefore, the summation over the intermediate states reduces to only two states close to the conduction band, *i.e.* we may adopt a four-band model where only four bands of interest are used.

In the following section, we shall treat expressions (2) and (3) by a procedure which allows explicitly only the intermediate states of interest.

### III. Geometrical and Dynamical Parts

The inner product  $\underline{\varepsilon}_i \cdot \underline{P}$  in cubic crystals can be written in the form (Koster *et al.* 1963, Hammermesh 1962):

$$\underline{\varepsilon}_i \cdot \underline{P} = \sum_r \underline{\varepsilon}_{ir}^{T*} \cdot \underline{P}_r^T, \quad i = 1, 2, 3, \quad (4)$$

where  $D^T$  is the irreducible representation (at  $\underline{k} = 0$ ) to which the vectorial operators  $\underline{P}$  and  $\underline{\varepsilon}_i$  belong.

Inserting (4) into (1) we obtain

$$\begin{aligned} \alpha(\omega_1) = c \sum_{f''} \sum_l \sum_{r, r', r''} \varepsilon_{1r}^{T*} \varepsilon_{2r'}^{T*} \varepsilon_{3r''}^{T*} \underline{M}_{f_0}^l \times \\ \{ (1 + \mathcal{P}_{12}) \Lambda(\omega_{12}) \Lambda(\omega_2) + (1 + \mathcal{P}_{23}) \Lambda(\omega_{23}) \Lambda(\omega_3) + \\ + (1 + \mathcal{P}_{13}) \Lambda(\omega_{13}) \Lambda(\omega_1) \}^2 \delta(E_{f''} - E_0 - \hbar\omega_1 - \hbar\omega_2 - \hbar\omega_3), \end{aligned} \quad (5)$$

where

$$\underline{M}_{f_0}^l = \langle f'' | \underline{P}_{r'}^T | \varphi_m^v \rangle \langle \varphi_m^v | \underline{P}_{r'} | \psi_n^\lambda \rangle \langle \psi_n^\lambda | \underline{P}_r^T | 0 \rangle.$$

In order to simplify expression (5) we use the Wigner-Eckart theorem in a form suitable for finite symmetry groups (Doni *et al.* 1974, Koster *et al.* 1963). For cubic groups we have

$$\langle \psi_n^\lambda | \underline{P}_r^T | 0 \rangle = \delta_{\lambda r} \delta_{nr} \langle \psi^\lambda | | \underline{P}^T | | 0 \rangle, \quad (6a)$$

$$\langle \varphi_m^v | \underline{P}_{r'}^T | \psi_n^\lambda \rangle = (vm | \pi', \lambda n) \langle \varphi^v | | \underline{P}^T | | \psi^\lambda \rangle, \quad (6b)$$

$$\langle f'' | \underline{P}_{r'}^T | \varphi_m^v \rangle = (\mu l | \pi'', vm) \langle f'' | | \underline{P}^T | | \varphi^v \rangle, \quad (6c)$$

and similarly the other two products involving  $r'$  and  $r''$ , where standard notations have been used to indicate the reduced matrix elements and Clebsch-Gordan coefficients (Hammermesh 1962).

Since throughout our treatment the only irreducible representations of interest are those at  $\underline{k} = 0$ , the Clebsch-Gordan coefficients of interest coincide with those of the crystal point groups, which are all tabulated in the work of Koster *et al.* (1963). Substituting (6) into (5) we obtain for the final forms:

$$\alpha(\omega_1) = c \sum_{f''} \{ G_\mu(\underline{\varepsilon}_1, \underline{\varepsilon}_2, \underline{\varepsilon}_3) | \underline{S} \underline{M}_{f_0}^l |^2 \}, \quad (7a)$$

where

$$G_\mu(\underline{\varepsilon}_1, \underline{\varepsilon}_2, \underline{\varepsilon}_3) = \sum_l \sum_r \varepsilon_{1r}^{T*} \varepsilon_{2r'}^{T*} \varepsilon_{3r''}^{T*} (\mu l | \pi, vm) |^2, \quad (7b)$$

$$\underline{M}_{f_0}^l = \langle f'' | | \underline{P}^T | | \varphi^v \rangle \langle \varphi^v | | \underline{P}^T | | \psi^\lambda \rangle \langle \psi^\lambda | | \underline{P}^T | | 0 \rangle, \quad (7c)$$

and

$$S = \Lambda(\omega_{23})[\Lambda(\omega_2) + \Lambda(\omega_3)] + \Lambda(\omega_{13})[\Lambda(\omega_1) + \Lambda(\omega_3)] + \Lambda(\omega_{12})[\Lambda(\omega_1) + \Lambda(\omega_2)], \quad (7d)$$

and we use the following property of Clebsch-Gordon coefficients:

$$(\nu m | \pi, \pi) = 1.$$

Expressions (7) show that in the case of cubic crystals it is always possible to separate the dynamical part, which is expressed in terms of reduced matrix elements of the momentum operator, from the geometrical part, which depends on the polarization vectors of the three photons. Eq. (7b) determines the polarization dependence for a given final state, while (7a) supplies useful information on the strength of contributions with different symmetry. Notice that these strengths are determined not only by electron-photon matrix elements, but also by the factors  $\Lambda(\omega_i)$  and  $\Lambda(\omega_{ij})$ .

#### IV. Application

To illustrate the results of the previous section, we consider the case of TlCl. The point group of TlCl is  $O_h$  and we use the notations of Koster *et al.* (1963) to label the irreducible representations. The momentum operator  $\underline{P}$  belongs to the irreducible representation  $\Gamma_4^-$ . The direct product  $\Gamma_4^- \times \Gamma_4^- \times \Gamma_4^-$  gives

$$\Gamma_4^- \times \Gamma_4^- \times \Gamma_4^- = \Gamma_1^- + \Gamma_2^- + 2 \Gamma_3^- + 2 \Gamma_4^- + 2 \Gamma_4^+ + \Gamma_5^- + 2 \Gamma_5^+. \quad (8)$$

For convenience the Clebsch-Gordon coefficients concerning the decomposition (8) are given in Table 1.

The geometrical factors  $G_{\mu}(\underline{\epsilon}_1, \underline{\epsilon}_2, \underline{\epsilon}_3)$ , corresponding to the product (8) defined in Eq. (7b), using Table 1, are given in the Appendix. The polarization dependence of the three-photon absorption coefficient becomes, in terms of these quantities

$$\alpha(\omega_1) \simeq |S|^2 \{G_{\Gamma_1^-} + G_{\Gamma_2^-} + 2 G_{\Gamma_3^-} + 2 G_{\Gamma_4^-} + G_{\Gamma_5^-} + 2 G_{\Gamma_5^+}\}. \quad (9)$$

To simplify expression (9), two particularly convenient geometrical configurations of light beams are used. In the parallel configuration we have:

$$\underline{\epsilon}_1 \equiv (\cos \theta, \sin \theta, 0); \quad \underline{\epsilon}_2 \equiv \underline{\epsilon}_3 \equiv (1, 0, 0).$$

In the  $\pi/4$  configuration we have

**Table 1.** The Clebsch-Gordan coefficients for the decomposition of the product representation  $\Gamma_4^- \times \Gamma_4^- \times \Gamma_4^-$  of the group  $O_h \cdot u_i$  ( $i = x, y, z$ ) and  $v_i$  ( $i = x, y, z$ ) denote two independent sets of functions transforming as the  $i^{\text{th}}$  row of  $\Gamma_4^-$  (Koster *et al.* 1963)

	$u_x v_x$	$u_x v_y$	$u_x v_z$	$u_y v_x$	$u_y v_y$	$u_y v_z$	$u_z v_x$	$u_z v_y$	$u_z v_z$
$\Gamma_1^\pm$	$\pm 1/\sqrt{3}$	0	0	0	$\pm 1/\sqrt{3}$	0	0	0	$\pm 1/\sqrt{3}$
$\Gamma_2^\pm$	$\pm 1/\sqrt{3}$	0	0	0	$\mp 1/\sqrt{3}$	0	0	0	$\pm 1/\sqrt{3}$
$\Gamma_3^\pm$	$\mp 1/\sqrt{6}$	0	0	0	$\mp 1/\sqrt{6}$	0	0	0	$\pm 2/\sqrt{6}$
	$\pm 1/\sqrt{2}$	0	0	0	$\mp 1/\sqrt{2}$	0	0	0	0
$\Gamma_4^\pm$	0	0	0	0	0	$\pm 1/\sqrt{2}$	0	$\mp 1/\sqrt{2}$	0
	0	0	$\pm 1/\sqrt{2}$	0	0	0	$\pm 1/\sqrt{2}$	0	0
	0	$\pm 1/\sqrt{2}$	0	$\mp 1/\sqrt{2}$	0	0	0	0	0
$\Gamma_5^\pm$	0	0	0	0	0	$\pm 1/\sqrt{2}$	0	$\pm 1/\sqrt{2}$	0
	0	0	$\pm 1/\sqrt{2}$	0	0	0	$\pm 1/\sqrt{2}$	0	0
	0	$\pm 1/\sqrt{2}$	0	$\pm 1/\sqrt{2}$	0	0	0	0	0

$$\underline{\varepsilon}_1 \equiv (\cos \theta, \sin \theta, 0); \underline{\varepsilon}_2 \equiv \underline{\varepsilon}_3 \equiv \left( \frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0 \right).$$

The values of  $G_\mu$  for these configurations are given in the Appendix. Then Eq. (9) for parallel and  $\pi/4$  configurations becomes, respectively,

$$\alpha_{\parallel}(\omega_1) \approx 1/3 |S|^2 (3 + \cos^2 \theta) \quad (10a)$$

and

$$\alpha_{\perp}(\omega_1) \approx 1/12 |S|^2 (25 - 3 \cos^2 \theta - 8 \sin 2\theta). \quad (10b)$$

Eq. (9) gives us individually the strength of electronic transitions to the different allowed final states of the proper symmetry by the photon absorption. Expressions (10) for the absorption rate gives the polarization dependence for two chosen configurations which are convenient for experimental observation.

## V. Experimental Situations

Three-photon absorption has been detected experimentally in alkali halides (Catalano *et al.* 1972) (NaCl, KCl, KI) by means of photoconductivity measurements. The exciting source was a Q-switched ruby laser and its second harmonics. A three-photon excited spontaneous luminescence in cubic ZnS is also reported (Catalano *et al.* 1973). The non-linear cross-section of the three-photon excitation with an Nd laser is obtained experimentally in CdS (Catalano *et al.* 1974) and GaP (Catalano *et al.* 1975).

In the above experiments, the authors were interested only in detecting the three-photon absorption rate and showing its behaviour as a function of the photon energy and laser intensity. Therefore, they were using unpolarized beams of light in their measurements. Unfortunately, no experimental data of the three-photon absorption with polarized photons are available.

However, it is of interest to use polarized photons to gain additional information. By using parallel and  $\pi/4$  geometrical configuration of the photon beams, in three-photon absorption measurements, one obtains explicitly the strength of the electronic transitions to the various allowed final states with the proper symmetry. This approach should be useful in the determination of the band structure of the crystal under consideration.

## VI. Conclusion

To conclude, we wish to emphasise the following results:

- 1) A simple procedure is given for investigating the polarization dependence of three-photon spectroscopy. The absorption rate is separated into two parts:
  - a) A dynamical part, which contains the allowed electron-photon matrix elements and the energy denominator, supplies the relative strength of the inter-band electronic transitions to all allowed final states with different symmetries.
  - b) A geometrical part, which depends on the polarization vectors of the three photons and gives the polarization dependence of the absorption rate.
- 2) From the above procedure, useful information about the band structure of the crystal can be gained, besides the usual information about the energy dependence already obtained earlier.
- 3) The case of TlCl has been discussed and an experimental method has been suggested by using two configurations polarized beam of photons.

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## Appendix

In this appendix we give the values of the geometrical part  $G_{\mu}(\underline{\epsilon}_1, \underline{\epsilon}_2, \underline{\epsilon}_3)$  for different symmetries of the final states and also its value for the two chosen geometrical configurations mentioned in the text:

$$G_{\Gamma_1^-}(\underline{\varepsilon}_1, \underline{\varepsilon}_2, \underline{\varepsilon}_3) = \left\{ -\frac{1}{\sqrt{3}} [(\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3x}) \right. \\ \left. + (\varepsilon_{1y}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1z}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1x}\varepsilon_{2z}\varepsilon_{3y}) \right. \\ \left. + (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3z} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3z} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3z}) \right\}^2,$$

$$G_{\Gamma_2^-}(\underline{\varepsilon}_1, \underline{\varepsilon}_2, \underline{\varepsilon}_3) = \left\{ \left[ -\frac{1}{\sqrt{3}} (\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3x}) \right. \right. \\ \left. \left. + \frac{1}{\sqrt{3}} (\varepsilon_{1y}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1z}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1x}\varepsilon_{2z}\varepsilon_{3y}) - \right. \right. \\ \left. \left. - \frac{1}{\sqrt{3}} (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3z} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3z} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3z}) \right]^2 \right\},$$

$$G_{\Gamma_3^-}(\underline{\varepsilon}_1, \underline{\varepsilon}_2, \underline{\varepsilon}_3) = \left[ \frac{1}{\sqrt{6}} (\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3x}) + \right. \\ \left. + \frac{1}{\sqrt{6}} (\varepsilon_{1y}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1z}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1x}\varepsilon_{2z}\varepsilon_{3y}) - \right. \\ \left. - \frac{2}{\sqrt{6}} (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3z} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3z} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3z}) \right]^2 + \\ + \left[ -\frac{1}{\sqrt{2}} (\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3x}) \right. \\ \left. + \frac{1}{\sqrt{2}} (\varepsilon_{1y}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1z}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1x}\varepsilon_{2z}\varepsilon_{3y}) \right]^2,$$

$$G_{\Gamma_4^\pm}(\underline{\varepsilon}_1, \underline{\varepsilon}_2, \underline{\varepsilon}_3) = \left[ \pm \frac{1}{\sqrt{2}} (\varepsilon_{1y}\varepsilon_{3x}\varepsilon_{3z} + \varepsilon_{1z}\varepsilon_{2y}\varepsilon_{3z} + \varepsilon_{1x}\varepsilon_{2z}\varepsilon_{3z}) \right. \\ \left. \mp \frac{1}{\sqrt{2}} (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3y}) \right]^2 \\ + \left[ \mp \frac{1}{\sqrt{2}} (\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3z} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3z} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3z}) \right.$$

$$\begin{aligned}
& \pm \frac{1}{\sqrt{2}} (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3x})^2 \\
& + [\pm \frac{1}{\sqrt{2}} (\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3y}) \\
& \pm \frac{1}{\sqrt{2}} (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3x})]^2, \\
G_{\Gamma_3^\pm} &= [\pm \frac{1}{\sqrt{2}} (\varepsilon_{1y}\varepsilon_{2x}\varepsilon_{3z} + \varepsilon_{1z}\varepsilon_{2y}\varepsilon_{3z} + \varepsilon_{1x}\varepsilon_{2z}\varepsilon_{3z}) \\
& \pm \frac{1}{\sqrt{2}} (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3y})]^2 + \\
& + [\pm \frac{1}{\sqrt{2}} (\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3z} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3z} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3z}) \\
& \pm \frac{1}{\sqrt{2}} (\varepsilon_{1z}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1x}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1y}\varepsilon_{2z}\varepsilon_{3x})]^2 + \\
& + [\pm \frac{1}{\sqrt{2}} (\varepsilon_{1x}\varepsilon_{2x}\varepsilon_{3y} + \varepsilon_{1y}\varepsilon_{2y}\varepsilon_{3y} + \varepsilon_{1z}\varepsilon_{2z}\varepsilon_{3y}) \\
& \pm \frac{1}{\sqrt{2}} (\varepsilon_{1y}\varepsilon_{2x}\varepsilon_{3x} + \varepsilon_{1z}\varepsilon_{2y}\varepsilon_{3x} + \varepsilon_{1x}\varepsilon_{2z}\varepsilon_{3x})]^2.
\end{aligned}$$

For Parallel configuration, the geometrical factor is equal to:

$$G_{\Gamma_1^-} = 1/3 \cos^2 \theta = G_{\Gamma_2^-},$$

$$G_{\Gamma_3^-} = 2/3 \cos^2 \theta,$$

$$G_{\Gamma_4^+} = G_{\Gamma_4^-} = 0,$$

$$G_{\Gamma_5^+} = G_{\Gamma_5^-} = 1/2 \sin^2 \theta.$$

For  $\pi/4$  configuration, the geometrical factor is equal to:

$$G_{\Gamma_1^-} = 1/12(4 - 3 \cos^2 \theta + 2 \sin 2 \theta) = G_{\Gamma_2^-},$$

$$G_{\Gamma_3^-} = 1/2(2 + \sin 2\theta),$$

$$G_{\Gamma_4^+} = 1/8(1 + 2 \cos^2 \theta - \sin 2\theta),$$

$$G_{\Gamma_5^+} = 1/8(4 - \cos^2 \theta + 2 \sin 2\theta).$$

### References

- Bader, T.R. and Gold, A.** (1968) Polarization dependence of two-photon absorption in solids, *Phys. Rev.* **171** : 997-1006.
- Badrysheva, A.I. and Moskalenko, S.A.** (1966) Symmetry of three-photon transitions, *Sov. Phys. Solid State* **8** : 2730-2739.
- Bassani, F.** (1972) *Multiphoton Transitions in Solids*, International School of Physics 'E. Fermi', Course L11. Academic Press, pp. 115-122.
- Bassani, F. and Hassan, A.R.** (1972) Two-photon transitions in solids, *Il Nuovo Cimento* **7** : 313-333.
- Catalano, I.M., Cingolani, A. and Minafra, A.** (1972) Photoconductivity of alkali halides, *Phys. Rev.* **B5** : 1629-1640.
- Catalano, I.M., Cingolani, A. and Minafra, A.** (1973) Two-photon luminescence in ZnS, *Phys. Rev.* **B8** : 1488-1493.
- Catalano, I.M., Cingolani, A. and Minafra, A.** (1974) Three-photon excitations in CdS, *Opt. Commun.* **11** : 254-258.
- Catalano, I.M., Cingolani, A. and Minara, A.** (1975) Four-photon transitions in ZnS, *Solid State Commun.* **16** : 417-424.
- Denisov, M.M. and Makarov, V.P.** (1973) Longitudinal and transverse excitations in semiconductors, *Phys. Status Solidi (b)* **56** : 9-23.
- Doni, E., Giralanda, R. and Pastiri Parravicini, G.** (1974) A note on the polarization dependence of two-photon absorption coefficient, *Phys. Status Solidi (b)* **65** : 203-215.
- Frohlich, D. and Stagninus, B.** (1967) New assignment of the band gap in the alkali bromides by two-photon spectroscopy, *Phys. Rev. Letters* **19** : 496-504.
- Frohlich, D., Stagninus, B. and Schonherr, E.** (1967) Two-photon absorption spectrum of CuCl, *Phys. Rev. Letters* **19** : 1032-1040.
- Frohlich, D., Stagninus, B. and Caps, T.** (1968) Symmetry assignment of electronic transitions of impurities in solids, *Solid State Commun.* **6** : 173-180.
- Frohlich, D., Stagninus, B. and Thurn, S.** (1970) Polarization dependence of the two-photon in CuCl, *Phys. Status Solidi* **40** : 287-294.
- Hammermesh, M.** (1962) *Group Theory and its Application to Physical Problems*, Addison Wesley, Reading, Mass.
- Inoue, M. and Toyozawa, Y.** (1965) Symmetry properties of the two-photon transitions, *J. Phys. Soc. Japan* **20** : 363-372.
- Koster, G.F.** (1958) Matrix element of the symmetry operator, *Phys. Rev.* **109** : 227-238.
- Koster, G.F., Dimmock, J.O., Wheeler, R.J. and Statz, H.** (1963) *Properties of the 32 Point Group*, MIT Press, Cambridge, Mass.
- Matsuoka, M.** (1967) Two-Photon Absorption in CdS, *J. Phys. Soc. Japan* **23** : 1028-1037.

- Park, K.** and **Stafford, R.G.** (1969) Evidence for an optical transition at a noncentrosymmetric point in KI, *Phys. Rev. Letters* **22** : 1426-1432.
- Pradere, F.** and **Mysyrowicz, A.** (1970) Polarization dependence of CdS by two-photon absorption, *Proceedings of the 10<sup>th</sup> International Conference on the Physics of Semiconductors*, Cambridge, Mass., pp. 243-251.
- Worlock, J.M.** (1972) Two-photon spectroscopy, in **Arecchi, F.T. and Schulz-Dubois, E.O.** (eds.) *Laser Handbook*, Vol. 2, North-Holland Publ. Co., Amsterdam, pp. 1313-1368.

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## الامتصاص الثلاثى الفوتونات المعتمد على الاستقطاب فى الجوامد

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١ - الجزء الهندسى الذى يعتمد على الاستقطاب المتجه للفوتونات الثلاثة .

٢ - الجزء الديناميكي والذى يحتوى على حاصل ضرب ثلاثة من كل من عناصر محدودة الإلكترون و الفوتون .

معلومات جديدة يمكن الحصول عليها بدراسة هذين الجزئين . البحث يحتوى على حالة كلوريد الثاليوم وكذلك اقتراح لتجربة عملية .

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