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Investigation of β-CdP_2 crystals by laser spectroscopy methods

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Abstract. CdP_2 single crystal of tetragonal modification is investigated by methods of laser-modulated spectroscopy at 293 K. The spectra of coherent two-photon absorption (TPA) have been measured and their theoretical analysis has been carried out. Double resonance of TPA at the total energy of two photons 2.60 eV is observed which occur through impurity level d3 in the gap at energy $E_c - 0.86$ eV (E_c is the energy of the conduction band edge). The time of transverse relaxation of electrons is found to be equal to $4.3 \cdot 10^{-14}$ s. The dependence is observed of single resonance on mutual orientation of vector of linear polarized light and crystallographic axis enabling us to conclude that the single TPA resonance in CdP_2 occurs between the states $\Gamma_7(\Gamma_4) \rightarrow \Gamma_7(\Gamma_3)$, $\Gamma_7(\Gamma_5) \rightarrow \Gamma_7(\Gamma_3)$, $\Gamma_6(\Gamma_5) \rightarrow \Gamma_6(\Gamma_1)$ in the case of the total energy of two photons is ranged from 2.5 to 3.11 eV. The energies of the states mentioned above are obtained.

Keywords: laser spectroscopy methods, band structure, two-photon absorption.

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1. Introduction

The results of a few studies of CdP_2 of tetragonal modification give foundations to make conclusion about the potentialities of this semiconductor compound in the field of optoelectronics and non-linear optics. From this the urgency is also raised of the investigation of two-photon absorption in CdP_2 by method of laser-modulated spectroscopy.

2. Experimental and discussion

The crystals investigated were grown by vapor phase technique in two-zone furnace. Pure cadmium and phosphorus constituents were taken in stoichiometric ratio for synthesis of single crystals. Samples had the configuration of rectangular bar with typical dimensions of $4\times4\times4$ mm^3 with optical axis oriented along Z-axis. Experimental set-up was the modification of that one described previously [l]. Unlike the set-up used earlier, in this set-up the portion of probe beam is directed into supplementary monochromator, after transmission of which it falls on the cathode of photo sensor. It allows us to determine the intensity of linear polarized radiation of probe beam in the case of the analyzer of polarized light is mounted before the monochromator.

In the spectroscopic method used information is extracted from measurements of intensity and polarization of probe wave with frequency ω_2 at excitation of media by pumping wave with frequency ω_1. Radiation of Q-switched Nd:YAG laser with the half-width of giant pulse $\tau_1 = 15$ ns was used as pumping wave, whereas radiation of powerful pulsed xenon lamp with pulse duration $\tau_2 = 150$ ns was used as probe wave. Both waves are propagated at the same direction. Due to high excitation of media by pumping wave the susceptibility of it is changed on the value of ΔK. This change is detected on frequency using probe beam. The quantities ΔK contain noncoherent and coherent contributions. Noncoherent contribution is attributed to resonant redistribution of state population, as coherent one is attributed to medium polariza-

tion. If impurity levels f with concentration N_f exist in the gap, then using the formalism of density matrix one can show that probe wave is affected by changing of level population via resonant susceptibility

$$\chi_R(\omega_2)=\frac{N_f}{\hbar}\left[\frac{\left|\left\langle c\left|e\cdot\vec{r}\right|f\right\rangle\right|^2}{\omega_2-\omega_{cf}+i\Gamma_{cf}}\Delta\rho_{fc}+\frac{\left|\left\langle f\left|e\cdot\vec{r}\right|v\right\rangle\right|^2}{\omega_2-\omega_{fv}+i\Gamma_{fv}}\Delta\rho_{vf}\right], \quad (1)$$

where $\Delta\rho_{fc}=\rho_{ff}-\rho_{cc}$ and $\Delta\rho_{vf}=\rho_{vv}-\rho_{ff}$ is the difference between population of states $|f\rangle$ and $|c\rangle$, $|v\rangle$ and $|f\rangle$, respectively. Due to fast relaxation of electrons and holes in c- and v-bands one obtains $\rho_{cc}=0$, $\rho=1$ and, consequently, $\Delta\rho_{fc}=\rho_{ff}$, $\Delta\rho_{vf}=1-\rho_{ff}$. In accordance with [1]

$$\rho_{ff}=\left(\rho_{ff}^{(0)}-\frac{\sigma'_{vf}}{\sigma'_{vf}+\sigma'_{fc}}\right)\times\exp\left(-\frac{\sigma'_{vf}+\sigma'_{fc}}{\hbar\omega_1}\cdot I(\omega_1)\cdot\tau_1\right)+\frac{\sigma'_{vf}}{\sigma'_{vf}+\sigma'_{fc}}, \quad (2)$$

where $I(\omega_1)$ is the intensity of pumping wave, $\rho_{ff}^{(0)}$ is the population of impurity level f before pumping, σ' is the cross-section of absorption of pumping wave. It follows from equations (1) and (2) that $\chi_R(\omega_2)$ exponentially depends on pumping intensity.

Coherent two-photon contribution is attributed to four-wave mixing caused by nonlinear susceptibility of the third order $\chi^{(3)}$. Microscopic expression for $\chi^{(3)}$ can be received by means of the perturbation theory. Cubic susceptibility may be written as the sum of resonant and non-resonant parts: $\chi^{(3)}=\chi_{NR}^{(3)}+\chi_R^{(3)}$. Coherent two-photon contribution is attributed to coherent resonant interaction caused by $\chi^{(3)}$. In consequence of such interaction two photons with frequencies $\omega_1(\vec{k}_1)$ and $\omega_2(\vec{k}_2)$ are absorbed coherently, whereas crystal is passed from initial $|v\rangle$ to final $|c\rangle$ states. This is the coherent TPA. TPA expressed in terms of altering of the intensity of probe wave $\Delta I(\omega_2)$ is proportional to the imaginary part of cubic susceptibility $\mathrm{Im}\chi^{(3)}$. $\Delta I(\omega_2)$ corresponds to changing of absorption coefficient of probe wave

$$\Delta K(\omega_2)=\frac{1}{d}\ln\frac{I(\omega_2)}{I(\omega_2)-\Delta I(\omega_2)}, \quad (3)$$

where $I(\omega_2)$ is the intensity of probe wave, d is the thickness of sample.

In consequence of four-wave mixing double refraction of probe beam is induced. This caused the rotation of polarization plane, and at the output of analyzer, crossed with the polarization plane of probe wave, intensity of probe beam increases [2]

$$\Delta I_y(\omega_2)\propto\left|\chi^{(3)}_{yxyx}+\chi^{(3)}_{yyxx}\right|^2 I_x^2(\omega_1)I_x(\omega_2) \quad (4)$$

in the case of linear polarization of the pumping wave, polarization plane of which is situated at angle of 45° with respect to the polarization plane of probe wave; and

$$\Delta I_y(\omega_2)\propto\left|\chi^{(3)}_{yxyx}-\chi^{(3)}_{yyxx}\right|^2 I^2(\omega_1)I_x(\omega_2) \quad (5)$$

in the case of circular polarization of the pumping wave. The angle θ of rotation of the polarization plane is defined by formula

$$\sin^2\theta=\frac{\Delta I_y(\omega_2)}{I_x(\omega_2)}. \quad (6)$$

In the case of both beams are propagated along the crystal symmetry axis for non-resonant part the ratio of symmetry is fulfilled $\left(\chi^{(3)}_{NR}\right)_{yxyx}=\left(\chi^{(3)}_{NR}\right)_{yyxx}$, however $\left(\chi^{(3)}_{R}\right)_{yxyx}\neq\left(\chi^{(3)}_{R}\right)_{yyxx}$ that is why one can receive

$$\Delta I_y(\omega_2)\propto\left|\left(\chi^{(3)}_{R}\right)_{yxyx}-\left(\chi^{(3)}_{R}\right)_{yyxx}\right|^2 I_x^2(\omega_1)I_x(\omega_2). \quad (7)$$

The important advantage of the method used consist of the fact, that special measures on maintenance of phase synchronism are not necessary; the condition $\vec{k}_2=\vec{k}_2+\vec{k}_1-\vec{k}_1$ is carried out identically. Therefore the direction of propagation of a probe beam with respect to a pumping beam can be arbitrary.

$\Delta K(\omega_2)$ consists of coherent and noncoherent parts. Noncoherent part $\Delta K^{(1)}(\omega_2)$ is attributed to the amplitude modulation of impurity single-photon absorption of probe wave, and coherent one equals to two-photon absorption coefficient $\Delta K^{(2)}(\omega_2)$:

$$\Delta K(\omega_2)=K(\omega_2)-K_0(\omega_2)=\left(K^{(1)}(\omega_2)+K^{(2)}(\omega_2)\right)-\left(K_0^{(1)}(\omega_2)+K_0^{(2)}(\omega_2)\right)=\Delta K^{(1)}(\omega_2)+K^{(2)}(\omega_2) \quad (8)$$

because of $K_0^{(2)}(\omega_2)=0$.

$$\Delta K^{(1)}(\omega_2)=4\pi\frac{\omega_2}{n_2c}\left|\left\langle\mathrm{Im}\,\chi^{(1)}(\omega_2)\right\rangle-\left\langle\mathrm{Im}\,\chi_0^{(1)}(\omega_2)\right\rangle\right|, \quad (9)$$

$$K^{(2)}(\omega_2)=4\pi\frac{\omega_2}{n_2c}\left\langle\mathrm{Im}\,\chi^{(3)}(\omega_2=\omega_1-\omega_1+\omega_2)\right\rangle\left|\vec{E}(\omega_1)\right|^2 \quad (10)$$

Accounting for in equation (9) expressions (1) and (2) one obtains exponential dependence of $\Delta K^{(1)}(\omega_2)$ on $I(\omega_1)$. $K^{(2)}(\omega_2)$ can belong to single- and double-resonance of TPA. At ordinary resonance the condition $\omega_1+\omega_2=\omega_{vc}$ is fulfilled, where ω_{vc} is the frequency of transition between v- and c-bands. In theory of such resonance virtual states are considered as intermediate. In the case of double resonance $\omega_1=\omega_{vf}$ and $\omega_2=\omega_{cf}$, or (and) $\omega_1=\omega_{cf}$ and $\omega_2=\omega_{fv}$ and real f levels are considered to be the in-

termediate states. Thus, $K^{(2)}(\omega_2)=K_1^{(2)}(\omega_2)+K_2^{(2)}(\omega_2)$, where $K_1^{(2)}(\omega_2)$ and $K_2^{(2)}(\omega_2)$ are the coefficients of single and double resonance of TPA respectively, and

$$\Delta K(\omega_2)=\Delta K^{(1)}(\omega_2)+K_1^{(2)}(\omega_2)+K_2^{(2)}(\omega_2). \qquad (11)$$

Each of these three spectroscopic components contain important spectroscopic information and they demand of particular analysis. For their separation kinetic, spectral, intensity, polarization and angle characteristics of $\Delta K(\omega_2)$ were investigated, analogous to those ones investigated in ZnP_2 previously [1].

In Fig. 1 the spectral dependencies $\Delta K(\omega_2)$ are shown at variable mutual orientation of the vectors of polarization and crystallographic axis. Also shown is kinetic characteristic $\Delta I(\omega_2)$, which is typical for long- and short-wavelength bands of spectra. The curves $K^{(2)}(\omega_2)$ were extracted from the dependencies $\Delta K(\omega_2)=f\left(I(\omega_1)\right)$ using extrapolation procedure [1] (by drawing straight lines through zero point in parallel to high-intensity parts of the dependencies analyzed). Resulting spectra $K^{(2)}(\omega_2)$ thus obtained are shown in Fig. 2. Above the dependence of $K^{(2)}(\omega_2)$ is shown on angle between the plain in which electric vectors $\vec{e}_1$ and $\vec{e}_2$ are confined and Z-axis.

At propagation of beams along the optical axis $\vec{c}$ of a crystal a spectrum $\sin^2\theta$ of polarized modulation spectroscopy was also measured. Its structure is similar to that one shown in Fig. 2 (curve number 6). Thus, the most convincing confirmation of the fact that the spectra shown in Fig. 2 belong to two-photon absorption phenomena is received.

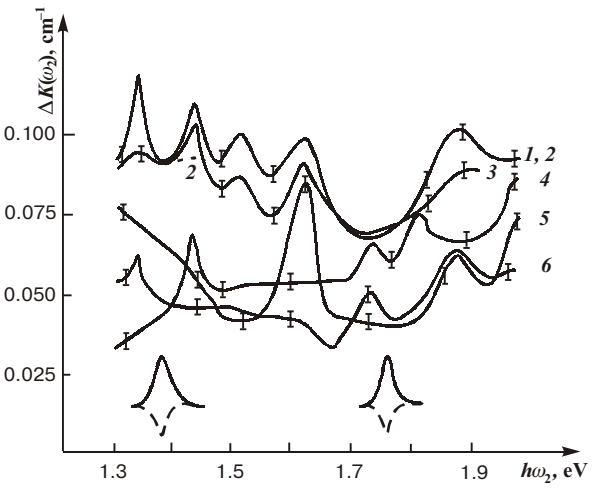

Fig. 1. Spectral dependencies of $\Delta K(\omega_2)$ in β-CdP_2:
$1-\vec{q}_1\|\vec{q}_2\|\vec{c}\perp\vec{e}_1\|\vec{e}_2$; $2-\vec{q}_1\|\vec{q}_2\|\vec{c}\perp\vec{e}_1\perp\vec{e}_2$; $3-\vec{q}_1\|\vec{q}_2\perp\vec{c}\perp\vec{e}_1\|\vec{e}_2$;
$4-\vec{q}_1\|\vec{q}_2\perp\vec{c}\|\vec{e}_1\|\vec{e}_2$; $5-\vec{q}_1\|\vec{q}_2\perp\vec{c}\perp\vec{e}_1\perp\vec{e}_2$; $6-\vec{q}_1\|\vec{q}_2\perp\vec{c}\|\vec{e}_1\perp\vec{e}_2$;
$\vec{q}_1,\vec{q}_2$ – wave vectors of laser and probe radiation; $\vec{e}_1,\vec{e}_2$ – principle vectors of electrical polarization of laser and probe radiation accordingly. Below the continuous lines represented are typical dependencies of $\Delta I(\omega_2)$ in long- and shortwave regions of the spectrum. The spectra correspond to points of maximum of kinetics. The dotted lines represent pulses of laser radiation. Intensity of laser radiation is 4 MW cm^{-2}.

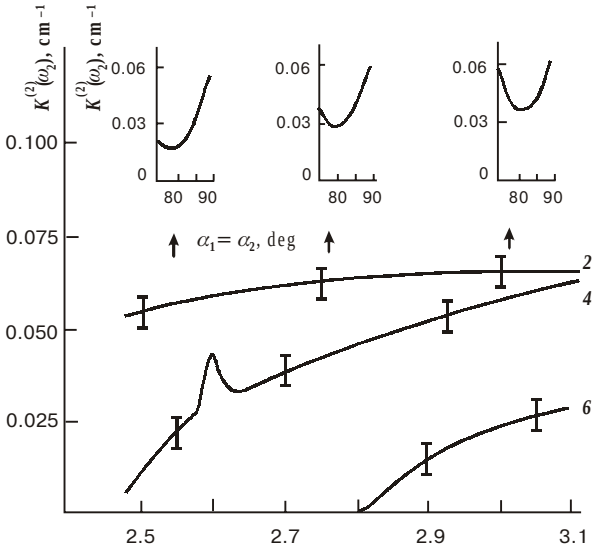

Fig. 2. Spectral dependencies of $K^{(2)}(\omega_2)$ in β-CdP_2. On mutual orientation of vectors the numbering of spectra corresponds to numbering in Fig. 1. Within measurement error limits, the spectra 1 and 3 not shown in this figure coincide with the spectrum 2, as well as the spectrum 5 with spectrum 6. Shown above are dependencies of $K^{(2)}(\omega_2)$ on the angle $\alpha_1=\vec{e}_1,\vec{c}$ $\vec{q}_1\|\vec{q}_2\perp c,\vec{e}_1\|\vec{e}_2$ at geometry of the experiment $\hbar\omega_1+\hbar\omega_2$ =2.54; 2.75; 3.01eV.

The spectra $\Delta K(\omega_2)$ (see Fig.1) contains several peaks with maximum at energies of photons of probe wave $\hbar\omega_2$ =1.34; 1.43; 1.50; 1.60; 1.69; 1.77; and 1.84 eV. Their intensity much depend on mutual orientation of vectors $\vec{e}_1$, $\vec{e}_2$ and $\vec{c}$. Comparing spectra $\Delta K(\omega_2)$ shown in Fig. 1 with spectra of TPA $K^{(2)}(\omega_2)$ shown in Fig. 2 one can see, that these peaks, except peak at 1.43 eV, belong to $\Delta K^{(1)}(\omega_2)$. In work [3] it has been shown, that peaks at energies 1.50; 1.60; 1,69 and 1.77eV are caused by optical transitions of electrons in donor-acceptor complexes, in which vacancies of Cd are acceptors and atoms of replacement of phosphorus, being in the 4-th, 3-rd, 2-th and 1-th coordination spheres are donors. The peak 1.84 eV was investigated in work [4] and is referred to intra-center transitions.

At propagation of beams along $\vec{c}$-axis, it has been found that the dependencies $K^{(2)}(\omega_2)$ on angle φ between $\vec{e}_1$ and $\vec{e}_2$ vectors as well as on the angle of rotation of a crystal around of its optical axis are absent. In the case of the propagation of beams is perpendicular to $\vec{c}$-axis distinct angular dependence of on angle between vectors $\vec{e}_1$ and $\vec{e}_2$ and dependence on angle between $\vec{e}_1\parallel\vec{e}_2$ and vector is observed (Fig. 2).

The peak number 4 shown on spectral dependence with a maximum at $\hbar(\omega_1+\omega_2)$ = 2.60 eV has the Lorenz contour, which is characteristic for double two-photon resonance. Coefficients of double TPA resonance $K_2^{(2)}(\omega_2)$ on deep impurity level f correspond to permit-permitted (p-p) two-photon transitions, as the deep local

electron states in the gap have not certain parity and transitions between them and *v*- and *c*-zones are dipole-permitted. For such transitions, in accordance with [1], in the case of $\sigma'_{fc}=0, \sigma'_{vf}\neq 0$,

$$K^{(2)}_{2,fc}(\omega_2)=\beta^m_{2,fc}I(\omega_1)\times$$
$$\times\left(1-\rho^{(0)}_{ff}\right)\exp\left(-\frac{\sigma'_{vf}}{\hbar\omega_1}\times I(\omega_1)\times\tau_1\right). \quad (12)$$

Here $\beta^m_{2,fc}$ is the constant of double resonance TPA, proportional to the maximum number N_f of intermediate levels participating in absorption:

$$\beta^m_{2,fc}\propto\frac{\Gamma_{cv}N_f}{\left[(\omega_1+\omega_2-\omega_{cv})+\Gamma^2_{cv}\right]}. \quad (13)$$

Γ_{cv} is attenuation constant. If $\sigma'_{fc}\neq 0, \sigma'_{vf}=0$, then

$$K^{(2)}_{2,vf}(\omega_2)=\beta^m_{2,vf}I(\omega_1)\times$$
$$\times\left(1-\rho^{(0)}_{ff}\right)\exp\left(-\frac{\sigma'_{fc}}{\hbar\omega_1}\times I(\omega_1)\times\tau_1\right). \quad (14)$$

Where equation (13) is carried out for $\beta^m_{2,fv}$, too. It is seen from (13) that the spectrum *p-p* of a double TPA resonance is represented by the narrow Lorenz line.

For uniaxial crystals such as β-CdP_2 with point-symmetry group C_{4v}, at propagation of beams along with $\vec{c}$-axis $K^{(2)}_2(\omega_2)$ depends on the angle between vectors $\vec{e}_1$ and $\vec{e}_2$ of linear polarization, and also on the type of beam polarization. In the case of linear polarization $K^{(2)}_{2,p-p}(\omega_2)\propto\cos(\vec{e}_1,\vec{e}_2)$. For circular polarization with opposite directed rotation $K^{(2)}_{2,p-p}(\omega_2)\neq 0$, and for circular polarization with identically directed rotation $K^{(2)}_{2,p-p}(\omega_2)=0$. These characteristics are typical for peak at 1.43 eV. $K^{(2)}_{2,p-p}(\omega_2)$ in β-CdP_2 is permitted-forbidden (*p-f*) type of transition [5], so angular and polarization dependencies obtained are not typical for it. The half-width of the resonant peak is $\hbar\Gamma_{cv}$ = 0.29 eV. From this value, the transverse relaxation time of electrons at a double TPA resonance $T_2=\frac{1}{\Gamma_{cv}}$ is obtained to be $4.3\cdot10^{-14}$ s. Intensity dependence of this peak is in accordance with Eq. (14), which corresponds to fulfilling the condition $\sigma'_{fc}\neq 0, \sigma'_{vf}=0$. It seems to be an evidence of impurity level of double resonance located at the depth more than 1.17 eV from the edge of valence band, instead of more than 1.17 eV from the edge of conductivity band and no more than 1.43 eV from the edge of the valence band. In this range of energy, the known level d_3 (E_c – 0,86) eV is located [6].

$K^{(2)}_1(\omega_2)$ as well as appropriate cubic susceptibility $\chi^{(3)}$ in general case is a tensor. The selection rule for them can be deduced from the theory of groups. They were received by Inoue and Toyozawa [7] for 32-point groups of crystal symmetry. The general view of the angular dependence $K^{(2)}_1(\omega_2)$ is determined by group *G* of crystal symmetry transformations. In a general case, the probability of TPA $W^{(2)}$ can be expanded on linearly independent invariants concerning symmetry transformations of group *G*, which are non-zero real values and which can be constructed of two vectors $\vec{e}_1, \vec{e}_2$, and two vectors $\vec{e}_1$* and $\vec{e}_2$*. Also $W^{(2)}=K^{(2)}_1(\omega_2)\times I(\omega_1)/\hbar\omega_2$.

Such expansion of $W^{(2)}$ for cubic symmetry was obtained in the work [8], and for hexagonal symmetry in [9]. Similarly, in agreement with the results of the work [7], it is possible to write down expansion of $W^{(2)}$ for crystals of tetragonal symmetry (point-symmetry group D_4) in the case of two linearly polarized beams of light:

$$W^{(2)}=\left[a_1\left(\vec{e}_{1z}\times\vec{e}_{2z}\right)+a_2\left|\left(\vec{e}_1\times\vec{e}_2\right)_z\right|\right]^2+$$
$$+\left[\tilde{a}_1\left|\left(\vec{e}_1\times\vec{e}^{\,*}_2\right)_x\right|\right]^2+\left[a_3\left(e_{1x}\cdot e_{2x}-e_{1y}\cdot e_{2y}\right)\right]^2+$$
$$+\left[a_4\left(e_{1z}\cdot|\vec{e}_{2\perp}|+|\vec{e}_{1\perp}|\cdot e_{2z}\right)\right]^2+ \quad (15)$$
$$+\left[a_5|\vec{e}_{1\perp}|\cdot|\vec{e}_{2\perp}|+\tilde{a}_2\left|\left(\vec{e}^{\,*}\times_1\vec{e}_2\right)_\perp\right|\cdot\right]^2.$$

Here Z-axis is directed along with axis C_4 and values $(a_i)^2$, which are proportional to intensities $I(\omega_1), I(\omega_2)$, depend on light frequencies ω_1, ω_2 as well as parameters of crystals, and do not depend on vectors $\vec{e}_1$, $\vec{e}_2$. The vectors cannot be determined by methods of the group theory. They can be calculated by the methods of the second order perturbation theory taking into account the light absorption probability for a real band structure of crystals.

The right part of the equation (15) contains a sum of five squares, which at symmetry transformation of crystal can be expanded on irreducible representations of group *D*4: $\Gamma_1, \Gamma_2, \Gamma_3, \Gamma_4, \Gamma_5$. These are the representations of transitions. They are expanded in direct products of irreducible representations of initial and final states of transitions. At linear polarization of light the transitions between states with different projections of a complete angular momentum of electrons on an axis of quantization, oriented along a pulse direction, are forbidden [9]. That is, transitions $\Gamma_6^{\pm}\to\Gamma_6^{\pm}$, $\Gamma_7^{\pm}\to\Gamma_7^{\pm}$, $\Gamma_5\to\Gamma_6^{\pm}, \Gamma_5\to\Gamma_7^{\pm}$, with amplitudes of dipole moments (equal to matrix elements of transitions), are transformed on irreducible representations Γ_1, and Γ_5, at symmetry transformation of group D_4. Therefore in the case of linear polarization of beams coefficients $\tilde{a}_1, a_3, a_4$ in (15) are equal to zero as well as $a_1, a_2, \tilde{a}_2, a_5$, are not.

3. Summary

The received theoretical results are shown in Table 1. They are in accordance with experimental results shown in Fig. 2 provided that . It is possible to explain equality by the fact of absence of transitions between states Γ6 (Γ_1, Γ_2) and in the Γ_6 (Γ_1, Γ_2) spectral region used, because of such states is absent, too [10].

Table 1.

Irreducible representations of dipole momentum of two-photon transitions	Irreducible representations of initial and final states of two-photon transitions	$\vec{q}_1 \| \vec{q}_2 \| [001]$, $\vec{e}_1(\cos\varphi_1, \sin\varphi_1, 0)$, $\vec{e}_2(\cos\varphi_2, \sin\varphi_2, 0)$, $\varphi_1 = \vec{e}_1, x, \varphi_2 = \vec{e}_2$, $x, \varphi = \vec{e}_1, \vec{e}_2$			$\vec{q}_1 \| \vec{q}_2 \| [010]$, $\vec{e}_1(\sin\alpha_1, 0, \cos\alpha_1)$, $\vec{e}_2(\sin\alpha_2, 0, \cos\alpha_2)$, $\alpha_1 = \vec{e}_1, z, \alpha_2 = \vec{e}_2, z, \alpha = \vec{e}_1, \vec{e}_2$						
		φ	0°	90°	α_1, degree α_2, degree α_3, degree	0 0 0	30 30 0	60 60 0	90 90 0	0 90 90	90 0 90
Γ1	$\Gamma_6^+(\Gamma_1) - \Gamma_6^+(\Gamma_1)$ $\Gamma_6^-(\Gamma_2) - \Gamma_6^-(\Gamma_2)$ $\Gamma_7^+(\Gamma_3) - \Gamma_7^+(\Gamma_3)$	$a_2^2 \sin^2\varphi$	0	a_2^2	0	0	0	0	0	0	0
	$\Gamma_7^+(\Gamma_4) - \Gamma_7^-(\Gamma_4)$	0	0		$(a_1 \cos\alpha_1 \cos\alpha_2)^2$	a_1^2	$\frac{9}{16}a_1^2$	$\frac{1}{16}a_1^2$	0	0	0
Γ2	$\Gamma_6^+(\Gamma_1) - \Gamma_6^-(\Gamma_2)$ $\Gamma_7^+(\Gamma_3) - \Gamma_7^-(\Gamma_4)$	0	0	0	0	0	0	0	0	0	0
Γ3	$\Gamma_7^+(\Gamma_3) - \Gamma_6^+(\Gamma_1)$ $\Gamma_7^-(\Gamma_4) - \Gamma_6^-(\Gamma_2)$	0	0	0	0	0	0	0	0	0	0
Γ4	$\Gamma_7^-(\Gamma_4) - \Gamma_6^+(\Gamma_1)$ $\Gamma_7^+(\Gamma_3) - \Gamma_6^-(\Gamma_2)$	0	0	0	0 0	0					
Γ5	$\Gamma_5(\Gamma_6^\pm + \Gamma_7^\pm) - \Gamma_6^\pm(\Gamma_1, \Gamma_2)$	a_5^2	a_5^2	a_5^2	$[a_5 \sin\alpha_1 \sin\alpha_2 +$ $+ \tilde{a}_2 \sin^2\alpha]$	0	$\frac{1}{16}a_5^2$	$\frac{9}{16}a_5^2$	a_5^2	0	0
	$\Gamma_5^+(\Gamma_3^\pm + \Gamma_7^\pm) - \Gamma_7^\pm(\Gamma_3, \Gamma_4^-)$	0	0	0		0	0	0	0	$\tilde{a}_2^2$	a_2^2
Number of the spectrum			1	2		4			3	6	5

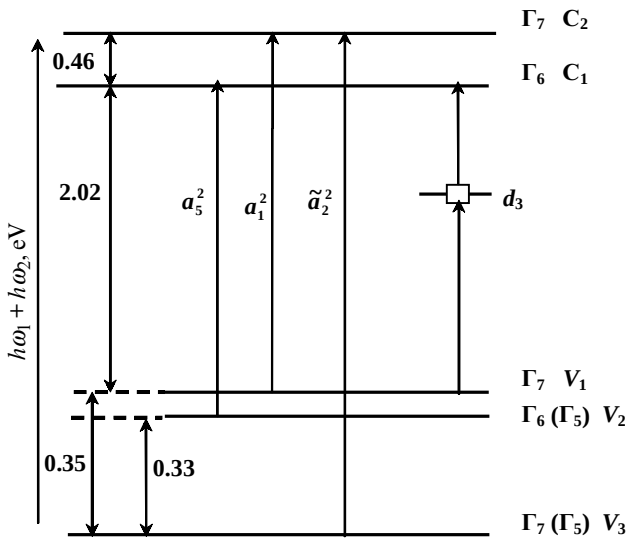

Fig. 3. The diagram of TPA for plane-polarized light in β-CdP_2.

The diagram of TWA appropriate to received results in is represented in Fig. 3, where and are amplitudes of TPA probabilities of an unary resonance, and d3 is the impurity level, on which double resonance comes true.

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