Optical second harmonic generation of oblique incident light in transmission in potassium dihydrogen phosphate crystal

V. Bhanthumnavin
Department of Physics and Institute of Science, Suranaree University of Technologies, Suranaree, Ampure Muang, Nakoranrajsima 30000, Thailand

Chi H. Lee
Department of Electrical Engineering, University of Maryland, College Park, Maryland 20742

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The intensity of the transmitted inhomogeneous and homogeneous second harmonic light generated from potassium dihydrogen phosphate (KDP) crystals immersed in an optically denser fluid 1-bromonaphthalene have been observed as a function of incident angle θ_i of the fundamental beam of a mode locked neodymium glass laser. The laser pulses have the polarization in [1$\bar{1}$0] direction with respect to the KDP crystallographic axes. The transmitted second harmonic generation in the neighborhood of critical angle of incidence is performed. The phase matching of second harmonic generation in transmission is demonstrated. Furthermore, we observed that no transmitted inhomogeneous and homogeneous second harmonic light at normal incident angle when the nonlinear polarization $\mathbf{P}^{\mathrm{NLS}}$ $(2w)$ lies in the direction [001] along the crystal face normal. The experimental result agrees well with the Bloembergen and Pershan theory.

I. INTRODUCTION

The theory of harmonic generation at the boundary of a nonlinear medium has been given by Bloembergen and Pershan (BP)[1] and has been verified experimentally for a variety of geometrical situations.[2–8] Nonlinear second harmonic generation in transmission has been studied extensively, however, most of the work has involved normal incidence with phase matching for maximum conversion efficiency. A wide variety of nonlinear interaction with oblique incident fundamental beam is not well investigated. This general situation of second harmonic generation (SHG), which has been theoretically analyzed by BP[1] three decades ago and more recently by Dick *et al.*,[9] provides a rich variety of nonlinear optical phenomena, which has not been experimentally demonstrated. The situation of nonlinear boundary condition is of particular interest. This work provides some experimental verification of the BP theory and those of Dick *et al.* It is shown that the two theoretical formalisms essentially predict the same result which agrees with the experimental observation.

It is the purpose of this paper to present a quantitative study of second harmonic generation in transmission with oblique incidence of the fundamental beam. The special emphasis is SHG in transmission in the vicinity of critical angles $\theta_{\mathrm{cr}}(w)$ and $\theta_{\mathrm{cr}}(2w)$ and conditions of null SHG in transmission with both normal and oblique incidence of the fundamental beam. It is for the first time that we observed null SHG from potassium dihydrogen phosphate (KDP) crystal at normal angle of incidence $\theta_i=0$. The results agree well with the prediction of both theories by BP and Dick *et al.*

In Sec. II, the BP theory is given in a form which will permit direct comparison with the experiment using uniaxial KDP crystal. Furthermore, criteria of null SHG in transmission is given for both normal and oblique incidence of fundamental beam. In Sec. III, the experimental arrangement is described and in Sec. IV, the experimental results are given for the case of SHG in transmission in the vicinity of critical angle of incidence. The analysis of null SHG in transmission at normal and oblique incidence and the condition of phase matching condition at oblique incidence are given with good agreement to experimental observation. Section V is the conclusion of the experimental results.

II. THEORY

The geometrical situation, just before total reflection occurs, is shown in Fig. 1. The fundamental beam is transmitted almost parallel to the surface in the nonlinear crystal KDP. According to BP theory,[1] there are reflected harmonic beams and two transmitted harmonic beams. The driven polarization wave propagates in the same direction as the transmitted laser beam. It has a wave vector $\mathbf{k}_S=2\,\mathbf{k}_L(w)$ and represents the particular solution of the inhomogeneous wave equation. In addition, there is the homogeneous solution with wave vector $\mathbf{k}_T(2w)$. In the nonlinear crystal KDP, the two transmitted harmonics beams are spatially distinct and readily observed separately. The relative magnitude of θ_S and θ_T in Fig. 1 depends on the magnitude of ordinary ray index of refraction $n_0(w)$ and extraordinary ray index of refraction $n_e(2w)$, respectively. If at particular crystallographic orientation of the KDP crystal such that $n_0^w > n_e^{2w}$, then $\theta_T > \theta_S$ or vice versa. As the angle of incidence θ_i in Fig. 1 is increased, it is intuitively clear that the beam with wave vector $\mathbf{k}_S$ $(2w)$ will disappear at the same time as the transmitted fundamental. The transmitted harmonic beam with wave vector $\mathbf{k}_T$ $(2w)$ will persist. There can be transmitted second harmonic

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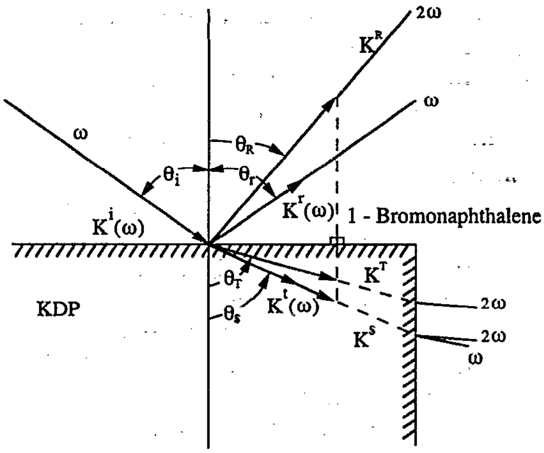

FIG. 1. K vectors of fundamental and second harmonic waves in the neighborhood of total reflection.

power even when the fundamental power is totally reflected. As θ_i becomes increasingly larger than the critical angle, this second harmonic wave will eventually disappear and only the reflected harmonic wave remains.

According to Fig. 1, the angles θ_R, θ_S, and θ_T of the reflected, inhomogeneous and homogeneous transmitted waves are given, respectively, by

$$n_{\text{liq}}(w)\sin\theta_i = n_{\text{liq}}(2w)\sin\theta_R = n(w)\sin\theta_S = n(2w)\sin\theta_T. \tag{1}$$

The refractive indices without subscripts refer to the KDP crystal. The components of nonlinear source polarization $\mathbf{P}^{\text{NLS}}$ along the cubic axes of the nonlinear KDP crystal are given in terms of the fundamental field components at each point inside the crystal by

$$P_z^{\text{NLS}}(2w) = \chi_{36}^{\text{NL}} E_x^T(w) E_y^T(w). \tag{2}$$

$P_x^{\text{NLS}}(2w)$, $P_y^{\text{NLS}}(2w)$ can be obtained by cyclic permutation of Eq. (2).

If the incident fundamental field is polarized perpendicular to the plane of incidence and along the $[1\bar{1}0]$ direction with respect to the crystallographic axes of KDP, the $\mathbf{P}^{\text{NLS}}$ will be along the [001] direction and lies in the plane of incidence. Equation (2) can be expressed in terms of the amplitude E_0 of the fundamental wave by

$$P_z^{\text{NLS}}(2w) = \chi_{36}^{\text{NL}} \eta (F_T^L E_0)^2, \tag{3}$$

where η is a geometrical factor that depends on the orientation of the fundamental field vector and nonlinear polarization component with respect to the crystallographic axes of the KDP. The linear Fresnel factor F_T^L describes the change of the amplitude of the fundamental wave on transmission at the crystal surface. If the linear polarization is perpendicular to the plane of incidence, it is given by

$$F_T^L = \frac{2\cos\theta_i}{\cos\theta_i + \sin\theta_{\text{cr}}(w)\cos\theta}. \tag{4}$$

The nonlinear polarization is the source of the three harmonic waves. The electric field amplitudes of the reflected and transmitted harmonic waves are given by

$$E_R(2w) = 4\pi P^{\text{NLS}} F_R^{\text{NL}}, \tag{5a}$$

$$E_S(2w) = 4\pi P^{\text{NLS}} F_S^{\text{NL}}, \tag{5b}$$

$$E_T(2w) = 4\pi P^{\text{NLS}} F_T^{\text{NL}}. \tag{5c}$$

The nonlinear Fresnel factors F_R^{NL}, F_S^{NL}, and F_T^{NL} have been calculated by BP theory.[1] For the case of second harmonic polarization $\mathbf{P}^{\text{NLS}}$ $(2w)$ parallel to the plane of incidence, the nonlinear Fresnel factors, according to the BP theory[1] becomes

$$F_{R,\parallel}^{\text{NL}} = \frac{\sin\theta_S \sin^2\theta_T \sin(\alpha+\theta_S+\theta_T)}{\epsilon_R(2w)\sin\theta_R \sin(\theta_T+\theta_R)\cos(\theta_T-\theta_R)\sin(\theta_S+\theta_T)}, \tag{6}$$

$$F_{S,\parallel}^{\text{NL}} = \frac{\sin\alpha}{\epsilon_s - \epsilon_T}, \tag{7}$$

$$F_{T,\parallel}^{\text{NL}} = -\frac{\epsilon_S^{1/2}\sin\alpha}{\epsilon_T^{1/2}(\epsilon_S-\epsilon_T)} + \frac{\epsilon_R^{1/2}}{\epsilon_T^{1/2}} F_{R,\parallel}^{\text{NL}}, \tag{8}$$

where $\epsilon_R^{1/2}(2w) = n_{\text{liq}}(2w)$; $\epsilon_S^{1/2} = n_{\text{KDP}}(w)$ and $\epsilon_T^{1/2} = n_{\text{KDP}}(2w)$. The angle α is the angle between the nonlinear polarization $\mathbf{P}^{\text{NLS}}$ $(2w)$ in the plane of incidence and the direction of the source vector $\mathbf{K}_S$. It is emphasized that these expressions remain valid in the case of total reflection.[4,5]

The time average second harmonic power carried by the harmonic beams is given by the real part of Poynting vector times the cross-sectional area A of the respective beams;

$$I_{R,S,T}(2w) = (c/8\pi)\epsilon_{R,S,T}^{1/2}(2w)\,|E_{R,S,T}(2w)|^2 A_{R,S,T}. \tag{9}$$

The $I_{R,S,T}(2w)$ in Eq. (9) is the intensity integrated over beam cross section, or power. This is the experimentally observed quantity. In the remainder of this paper, intensity and power will be used interchangeably in similar manner as in Refs. 4–6. $A_{R,S,T}$ in Eq. (9) is the cross-sectional area of the beam and is given by

$$A_{R,S,T} = (dd'\cos\theta_{R,S,T})/\cos\theta_i, \tag{10}$$

where dd' is the rectangular slit which defines the size of the incident laser beam. By substitution of relevant expression into Eq. (9), we finally obtain

$$I_R(2w)=(c/8\pi)\epsilon_R^{1/2}|E_0|^4 dd'(4\pi\chi_{36}^{\mathrm{NL}})^2\eta^2|F_L|^4|F_{R,\parallel}^{\mathrm{NL}}|^2 \times\cos\theta_R(\cos\theta_i)^{-1}, \quad (11)$$

$$I_S(2w)=(c/8\pi)\epsilon_S^{1/2}|E_0|^4 dd'(4\pi\chi_{36}^{\mathrm{NL}})^2\eta^2|F_L|^4|F_{S,\parallel}^{\mathrm{NL}}|^2 \times\cos\theta_S(\cos\theta_i)^{-1}, \quad (12)$$

$$I_T(2w)=(c/8\pi)\epsilon_T^{1/2}|E_0|^4 dd'(4\pi\chi_{36}^{\mathrm{NL}})^2\eta^2|F_L|^4|F_{T,\parallel}^{\mathrm{NL}}|^2 \times\cos\theta_T(\cos\theta_i)^{-1}, \quad (13)$$

for the intensity of reflected, transmitted source, and transmitted homogeneous harmonic waves, respectively. The expression of Eqs. (12) and (13) will facilitate to obtain the theoretical curves of intensity of harmonic waves and be compared to experimental data.

When the two transmitted second harmonic beams are not spatially resolved, the sum of homogeneous and inhomogeneous intensities $I_{\mathrm{total}}(2w)=I_S(2w)+I_T(2w)$ is observed. The total transmitted second harmonic intensity $I_{\mathrm{total}}(2w)$ is equal to the average intensity of the interference pattern of the two transmitted beams, which is observed in the more common geometry that the nonlinear crystal is a plane-parallel platelet and the light beams are nearly normal incidences. However, for a wedge-shaped sample, as in the case under study here, a spatial average is taken over the interference pattern in the direction normal to the surface of entry, where the two transmitted beams overlapped.[3,4] Therefore, the total transmitted harmonic intensity is equal to the sum of the intensities of the separated harmonic beams.

The uniaxial KDP crystal is employed for transmitted SHG at phase matching condition by birefringence. The phase matching angle θ_m is the result by making the birefringence $(n_0^{2w}-n_e^{2w})$ equal to the dispersion $(n_0^{2w}-n_0^{w})$ at the phase matching angle θ_m, which as the consequences will give $n^{2w}=n_0^{w}$ at this condition. Furthermore, the determination of $n_e^{2w}(\theta)$ for a specific value of θ can be obtained from the equation of index ellipsoid, given by

$$\frac{1}{[n_e^{2w}(\theta)]^2}=\frac{\cos^2\theta}{[n_0^{2w}]^2}+\frac{\sin^2\theta}{[n_e^{2w}(\pi/2)]^2}. \quad (14)$$

A. Criteria of null transmitted SHG

According to the BP theory for the case of $\mathbf{P}^{\mathrm{NLS}}$ $(2w)$ lying in the plane of incidence, the nonlinear Fresnel factors in reflection and transmission are given by Eqs. (6), (7), and (8), respectively. As a consequence, the intensity of reflected, transmitted in homogeneous and homogeneous harmonic beams are given by Eqs. (11), (12), and (13), respectively. Analysis of Eqs. (6), (7), and (8) derived from the BP theory leads us to the conditions of null transmitted second harmonic intensities. These conditions can be summarized as follows.

(1) For the case of angle of incidence θ_i in the neighborhood of critical angle $\theta_{\mathrm{cr}}^{(w)}$ and $\theta_{\mathrm{cr}}^{(w)}$. Under this condition one can obtain transmitted homogeneous and inhomogeneous intensities $I_T(2w)=0$ and $I_S(2w)=0$. This means there is neither homogeneous nor inhomogeneous intensities at all, when $\theta_i>\theta_{\mathrm{cr}}^{(2w)}$ and $\theta_i>\theta_{\mathrm{cr}}(w)$, respectively. Since at total internal reflection the terms θ_T and θ_S become imaginary. Therefore, $I_T(2w)$ and $I_S(2w)$ become zero and in our case this situation is confirmed by experimental results as indicated in Fig. 3.

(2) Furthermore, the condition of $I_S(2w)=0$ and $I_T(2w)=0$ can occur aside from the total reflection case. The situation can be achieved when $\alpha=0°$, $\theta_S=0°$, and $\theta_T=0°$. Under this condition the fundamental beam has the angle of normal incidence ($\theta_i=0°$) and the $\mathbf{P}^{\mathrm{NLS}}$ $(2w)$ must be along the face normal of the crystal which is the optical axis in Z direction of [001] as indicated in the inset of Fig. 4.

When $\alpha=0°$, $\theta_S=0°$, and $\theta_T=0°$, nonlinear Fresnel factors $F_{R,\parallel}^{\mathrm{NL}}$, $F_{S,\parallel}^{\mathrm{NL}}$, and $F_{T,\parallel}^{\mathrm{NL}}$ in Eqs. (6)–(8) become zero and as the consequences $I_R(2w)$, $I_S(2w)$, and $I_T(2w)$ also become zero, respectively. It is interesting to notice under this condition not only inhomogeneous and homogeneous harmonic intensities become zero, but also reflect harmonic intensity in addition to the condition of the nonlinear Brewster angle.[1,6,9] Therefore, no second harmonic generation occurs at normal incidence for the crystal that has particular crystallographic orientation as indicated in Fig. 4. The physical interpretation of null transmitted second harmonic inhomogeneous, homogeneous, and reflected intensities is that the nonlinear polarization $P^{\mathrm{NLS}}(2w)$ cannot radiate inside the medium in the direction which otherwise would yield transmitted inhomogeneous, homogeneous, and reflected harmonic rays, respectively. This situation is an uncommon phenomenon that one cannot obtain transmitted second harmonic light in transmission from nonlinear optical medium as compared to the simple normal incidence case. Thus, it has been demonstrated for the first time the null of $I_S(2w)$ and $I_T(2w)$ and they agree well to the prediction of the BP theory.

(3) For the case of $\alpha=0°$, $\theta_S>0°$ and $\theta_T>0°$. In general for a birefringence crystal, e.g., KDP, $\theta_S\neq\theta_T$ except at phase matching condition. Therefore, according to Eq. (8), one will find that $F_{T,\parallel}^{\mathrm{NL}}$ will never become zero at this condition. However, at this condition where $\alpha=0°$ and $\theta_S>0°$, the $F_{S,\parallel}^{\mathrm{NL}}$ in Eq. (7) becomes zero. This means that the transmitted inhomogeneous second harmonic intensity $I_S(2w)=0$. The corresponding situation inside the medium is that the vector $\mathbf{K}^S$ is parallel to the $\mathbf{P}^{\mathrm{NLS}}$ $(2w)$, which cannot radiate inside the medium in the direction which would otherwise yield an inhomogeneous transmitted ray. It is important to point out that the explanation is not applicable to the case of having $\mathbf{K}^T$ parallel to $\mathbf{P}^{\mathrm{NLS}}$ $(2w)$, where one expects to obtain $I_T(2w)=0$. When $\mathbf{K}^T$ is parallel to $\mathbf{P}^{\mathrm{NLS}}$ $(2w)$, we will have $\alpha\neq0°$ and $\theta_S\neq\theta_T$ as indicated earlier. Hence, $F_{T,\parallel}^{\mathrm{NL}}$ in Eq. (8) will not become zero. Therefore, the condition of $\alpha=0°$, $\theta_S>0°$ and $\theta_T>0$ yields only zero intensity of transmitted inhomogeneous second harmonic wave. Furthermore, the total transmitted second harmonic intensity, $I_{\mathrm{total}}(2w)=I_S(2w)+I_T(2w)$

will never become zero. However, $I_{total}(2w)$ will be extremely low and will possess a "dip" at the angle of incidence θ_i related to this condition. This situation is demonstrated by an experimental result indicated in Fig. 6.

III. EXPERIMENTAL TECHNIQUE

A. KDP crystal preparation

The nonlinear crystal used in the experimental KDP (potassium dihydrogen phosphate) crystal of several crystallographic cuts as indicated in the experimental results section. KDP crystal is transparent at the fundamental and second harmonic wavelengths, respectively. This will enable an investigation in transmission. Furthermore, its linear index of refraction is relatively low so that total reflection from it is possible via the optically denser linear fluid 1-bromonaphthalene. Typical dimension of the KDP crystal is $15\times25\times8$ mm^3. The entrance and exit surfaces were polished optically flat to $\lambda/5$ at the *D*-line of sodium light and none of the surfaces were coated. The refractive indices of the KDP crystal at the fundamental and second harmonic frequencies are

$$n_0^w=1.4943; \quad n_0^{2w}=1.5130; \quad n_e^{2w}=1.4708.$$

The crystal was mounted on the aluminum holder which was connected to an angular rotational device mounted on a platform above the liquid cell. The variation of the angle of incidence θ_i was performed by the rotational device which had the axis of rotation tangential to the incident surface of the crystal. The crystal could be positioned with an accuracy of 0.01°.

B. Optically dense fluid

The KDP crystal was immersed in the optically denser fluid 1-bromonaphthalene which has larger indices than the crystal at both frequencies w and $2w$. The fluid is transparent in the range of wavelength of 0.4–1.6 μm. The indices of refraction of the fluid at fundamental and harmonic frequencies are $n_{liq}(w)=1.6262$ and $n_{liq}(2w)=1.6701$, respectively. From Eq. (1), we found the critical angles for total reflection for fundamental and second harmonic frequencies to be $\theta_{cr}(w)=66.78°$ and $\theta_{cr}(2w)=64.76°$, respectively.

C. Optical arrangement

The laser used in the experiment is a standard *Q*-switched mode locked Nd:glass laser system with Brewster cuts at both ends of the Nd:glass rod as shown in Fig. 2. The fundamental beam was linearly polarized by means of Glann Kappa prism. To ensure the fundamental beam be polarized along the $[1\bar{1}0]$ direction with respect to the crystallographic axes of KDP crystal, a half-wave plate was used to rotate the electric field of the laser beam from horizontal to vertical polarization. The fundamental beam before entering the liquid cells was regulated by a rectangular slit of 1×5 mm^2. The liquid cell had a hexagonal shape with six circular fused quartz windows. The detecting system of SHG from KDP crystal was mounted on an aluminum arm pivoted underneath the liquid cell. The axis of rotation of the arm was common to the axis of the crystal rotation which was the line tangential to the incident surface of the KDP. The second harmonic signal was isolated from the fundamental by the standard spectral filtering technique as indicated in Fig. 2. A slit of 4×10 mm^2 situated 50 cm away from the liquid cell was employed for the separation of transmitted inhomogeneous harmonic beam from the transmitted homogeneous beams when the investigation of transmitted SHG near the critical angle of incidence $\theta_{cr}(w)$. In this region, the two transmitted homogeneous and inhomogeneous harmonic beams are separated and vanished at the critical angle of incidence $\theta_{cr}(2w)$ and $\theta_{cr}(w)$, respectively. However, for the investigation of transmitted SHG for angle of incidence θ_i being smaller and far from the neighborhood of $\theta_{cr}(w)$ especially for the range of $0°<\theta_i<50°$, where the two transmitted beams are not completely spatially resolved and the investigation is involving the total transmitted harmonic intensity, the $I_{total}(2w)$ was obtained by placing the slit closer to the liquid cell and a biconvex lens was employed in front of a photomultiplier. Each data point was normalized by the monitor intensity in order to eliminate the effect caused by the laser intensity fluctuation.

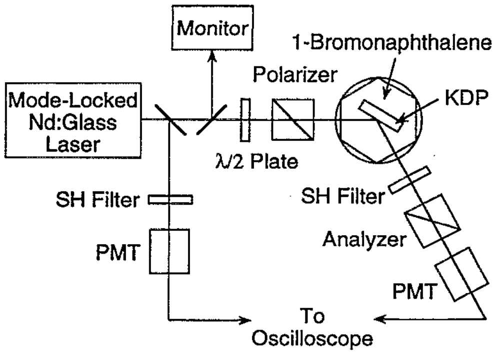

FIG. 2. Experimental arrangement for measuring transmitted SHG.

IV. EXPERIMENTAL RESULTS

A. Transmitted homogeneous and inhomogeneous second harmonic generation in the neighborhood of critical angles

The KDP crystal in the experiment has dimension of $25\times15\times8$ mm^3 and is immersed in the optically denser liquid 1-bromonaphthalene. The crystal has its face normal in the [001] direction (optic axis) and the fundamental laser beam is polarized along the $[1\bar{1}0]$ direction which is normal to the plane of incidence as indicated in Fig. 3. In this situation, the fundamental beam is transmitted almost parallel to the surface of KDP crystal. According to the BP theory,[1] there are two transmitted harmonic beams.

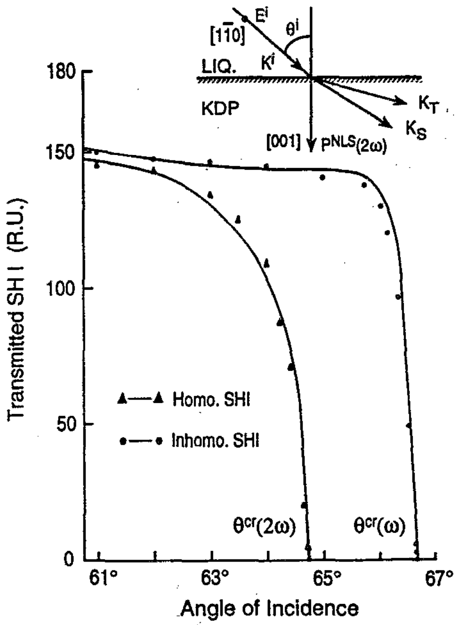

FIG. 3. The intensities of inhomogeneous and homogeneous transmitted second harmonic waves in the neighborhood of critical angle $\theta^{cr}(w)$ and $\theta^{cr}(2w)$.

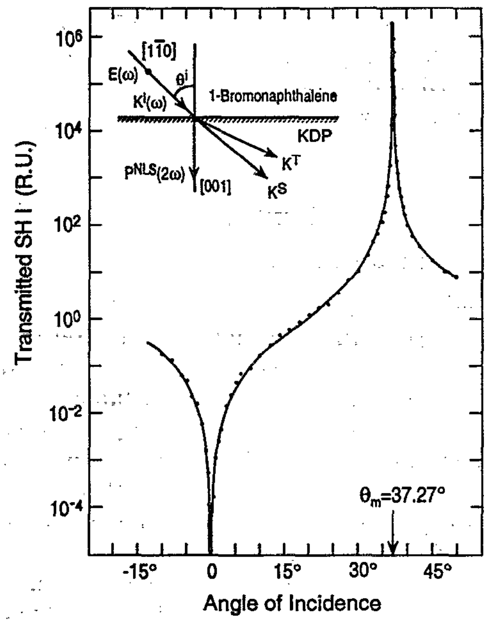

FIG. 4. The total transmitted second harmonic intensity $I_{\text{total}}(2w)=I_S(2w)+I_T(2w)$ as a function of angle of incidence θ_i. $I_S(2w)=I_T(2w)=0$ at $\theta_i=0$ and phase matching angle $\theta_i^m=37.27°$.

The driven polarization wave propagates the same direction as the transmitted laser beam. It has a wave vector $\mathbf{K}_S=2\ \mathbf{K}_{\text{laser}}(w)$. Furthermore, there is a homogeneous harmonic wave with a wave vector $\mathbf{K}_T(2w)$. In the experiment, the KDP crystal has right angular corners and the two transmitted harmonic beams are spatially distinct and readily observed separately. According to the KDP crystallographic orientation as indicated in Fig. 3 and also the investigation of transmitted SHG in the neighborhood of critical angles, the KDP crystal employed in the experiment under this situation has the value of $n_0^w > n_e^{2w}(\theta)$. Therefore, in the neighborhood of critical angle, for a given angle of incidence θ_i there exist θ_S and θ_T which can be determined by Eq. (1), such that $\theta_T > \theta_S$. Therefore, from the experiment it is found that $\theta_{\text{cr}}(w) > \theta_{\text{cr}}(2w)$. It is clear that the beam with wave vector $\mathbf{K}_T(2w)$ will disappear first at $\theta_i=\theta_{\text{cr}}(2w)$ and the ray with wave vector $\mathbf{K}_S$ will disappear at the same time as the fundamental beam. As the angle of incidence θ_i becomes larger than $\theta_{\text{cr}}(w)$ there is no transmitted second harmonic beam.

The inhomogeneous and homogeneous harmonic intensities are given by Eqs. (11) and (12), respectively. The drawn solid curves in Fig. 3 are theoretical curves predicted by the BP theory and are calculated from the last four factors, respectively,

$$|F^L|^4 |F_{S,T}^{\text{NL}}|^2 \cos\theta_{S,T}(\cos\theta_i)^{-1}.$$

The experimental dotted points are in excellent agreement to the theoretical prediction that the homogeneous and inhomogeneous harmonic intensities will be terminated at $\theta_{\text{cr}}(2w)$ and $\theta_{\text{cr}}(w)$, respectively. The reason that the homogeneous and inhomogeneous harmonic intensities vanish at $\theta_{\text{cr}}(2w)$ and $\theta_{\text{cr}}(w)$, respectively, is that when θ_i is greater than $\theta_{\text{cr}}(2w)$ and $\theta_{\text{cr}}(w)$ the values of $\cos\theta_T$ and $\cos\theta_S$ become purely imaginary, respectively. As a consequence, the two harmonic intensities will become imaginary which are not physically allowed. It is worthwhile to notice that, from Fig. 3, there is an interval $\theta_{\text{cr}}(2w) < \theta_i < \theta_{\text{cr}}(w)$ which is about 2.02° interval. In this region there exists only inhomogeneous harmonic intensity which has direct association with the nonlinear polarization $\mathbf{P}^{\text{NLS}}$ $(2w)$ and with the nonlinear susceptibility χ^{NL}. The knowledge of inhomogeneous second harmonic generation in this particular region will directly facilitate the study of $\mathbf{P}^{\text{NLS}}$ $(2w)$ and χ^{NL} of a nonlinear medium.

B. Null SHG in transmission

In this experiment KDP crystallographic orientation is selected such that at the normal incidence of the fundamental beam, there is no harmonic generation, neither in transmission nor in reflection. The KDP crystal used in the experiment was immersed in the optically denser fluid 1-bromonaphthalene and has its optic (z) axis lying along the face normal as indicated in Fig. 4. The fundamental beam was polarized along the [0$\bar{1}$0] direction with respect to the KDP crystallographic axes. Thus the $\mathbf{P}^{\text{NLS}}$ $(2w)$ was along the [001] direction which was the face normal direction. From the experiment, we found that at normal incidence where $\theta_i=0°$, there are no inhomogeneous and homogeneous second harmonic signals at all. This means the $I_S(2w)=0$ and also $I_T(2w)=0$ at this angle of incidence. The experimental data indicated in Fig. 4 agree well with the prediction of the BP theory. The null inhomogeneous and homogeneous harmonic intensities in this case can be explained as follows. At normal incidence we have $\theta_i=0°$, $\theta_S=0°$, $\theta_T=0°$, and also $\alpha=0°$. Therefore, according to

Eqs. (6)–(8), the $F^{NL}_{R,\|}$, $F^{NL}_{S,\|}$, and $F^{NL}_{T,\|}$ become zero, respectively, and as a consequence $I_R(2w)$, $I_S(2w)$, and $I_T(2w)$ given by Eqs. (11)–(13) all become zero. It is worthwhile to indicate that not only two transmitted harmonic intensities become zero, but also in this case the reflected second harmonic intensity $I_R(2w)=0$. Thus for the first time we have demonstrated that for a certain particular crystallographic orientation of a nonlinear medium, there can be no second harmonic generation in both transmission and reflection at normal incidence. The physical interpretation of null transmitted inhomogeneous, homogeneous, and reflected harmonic intensities, respectively, is due to the fact that nonlinear polarization $\mathbf{P}^{NLS}(2w)$ cannot radiate inside the medium in the direction which otherwise would yield transmitted inhomogeneous, homogeneous, and reflected harmonic rays. In other words in terms of the dipole radiation point of view, the nonlinear polarization $\mathbf{P}^{NLS}(2w)$ cannot radiate harmonic waves in the direction of its oscillation. The experimental result agrees well with the BP theory.[1]

Furthermore, it is worthwhile to point out that the experimental result at normal angle of incidence, as indicated in Fig. 4 also agrees well to the prediction of null transmitted second harmonic intensity $I^T_P(2w)$ given by Dick *et al.*[9] According to Dick *et al.* the transmitted second harmonic intensity is given by

$$I^T_P(2w) \simeq |\tilde{f}^T_X P^{NLS}_X + \tilde{f}^T_Z P^{NLS}_Z|^2, \tag{15}$$

where $I^T_P(2w)$ is the transmitted second harmonic intensity due to the nonlinear polarization $\mathbf{P}^{NLS}(2w)$ lying parallel (P) to the plane of incidence. $\tilde{f}^T_X$ and $\tilde{f}^T_Z$ are X and Z components of nonlinear Fresnel factors. From Eq. (15), one can obtain

$$I^T_P(2w) \simeq \left| \frac{dw_3 \sin\theta_T}{\sin(\theta_R+\theta_T)\cos(\theta_T-\theta_R)} (\cos\theta_R P^{NLS}_X + \sin\theta_R P^{NLS}_Z) \right|^2, \tag{16}$$

where $w_3=w_1+w_2=2w$ (for second harmonic generation) and (X,Y,Z) are crystal surface axes systems. According to Fig. 5,

$$P^{NLS}_X = P^{NLS}_\| \sin(\alpha+\theta_S),$$
$$P^{NLS}_Z = P^{NLS}_\| \cos(\alpha+\theta_S). \tag{17}$$

By substitution P^{NLS}_X, P^{NLS}_Z in Eq. (17) into Eq. (16) one obtains

$$I^P_T(2w) \alpha \left| \frac{P^{NLS}_\| \sin\theta_T \sin(\alpha+\theta_S+\theta_R)}{\sin(\theta_T+\theta_R)\cos(\theta_T-\theta_R)} \right|^2. \tag{18}$$

Therefore, the condition for $I^T_P(2w)=0$ is

$$\alpha+\theta_S+\theta_R=0,\pi \tag{19}$$

or

$$\alpha+\theta_S+\theta_R=0; \quad \alpha+\theta_S=-\theta_R. \tag{20}$$

For the case of transmitted second harmonic intensity at normal incidence, where $\theta_i=0°$ and $\theta_S=\theta_T=\theta_R=0°$, the condition for null transmitted second harmonic intensity given by Eq. (20) is satisfied. This is agreeable to the prediction of Dick *et al.*[9] and the experimental result is shown in Fig. 4. Furthermore, for the case of $\theta_i \neq 0°$, we have the condition for null transmitted second harmonic intensity given by Eq. (20), where $\alpha+\theta_S=-\theta_R$. The angle $\alpha+\theta_S$ is the angle between $\mathbf{P}^{NLS}(2w)$ and the face normal. The condition, where $\alpha+\theta_S=-\theta_R$, can be depicted as indicated in Fig. 5. The wave vector $\mathbf{K}^{-R}$ is in the same direction of $\mathbf{P}^{NLS}(2w)$. Therefore, the $\mathbf{P}^{NLS}(2w)$ cannot radiate inside the medium in the direction which would otherwise yield an inhomogeneous refracted second harmonic ray.

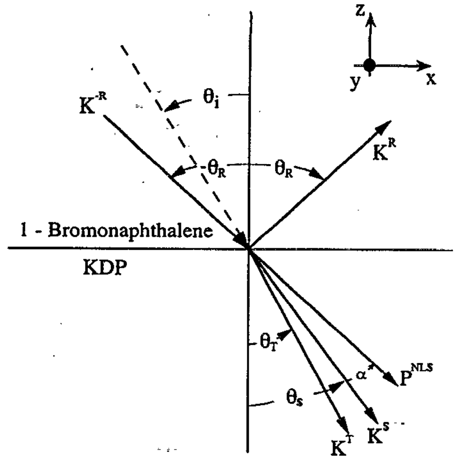

FIG. 5. Null transmitted inhomogeneous second harmonic generation where $\alpha+\theta_S+\theta_R=0$. The PNLS$(2w)$ source cannot radiate in the medium in the direction $\mathbf{K}^{-R}$ which would otherwise yield transmitted inhomogeneous second harmonic wave.

In addition, the experimental set up with the crystallographic cut of KDP crystal indicated in Fig. 4 will provide the phase matching SHG in transmission. The phase matching angle of KDP crystal used in the experiment is $\theta_m=41.2°$ inside the crystal and away from the optic (Z) axis which in this case is the face normal direction of the crystal. Thus by using Eq. (1), the angle of θ_i corresponding to θ_m is equal to $\theta_i^m=37.27°$.

The total transmitted second harmonic intensity is the sum of homogeneous and inhomogeneous intensities

$$I_{total}(2w)=I_S(2w)+I_T(2w), \tag{21}$$

where $I_S(2w)$ and $I_T(2w)$ are given by Eqs. (12) and (13), respectively. The theoretical curve shown in Fig. 4 was computed from the sum of the last four factors of Eqs. (12) and (13) and it can be expressed as

$$I_{total}(2w) \simeq |F^L_T|^4 (\cos\theta_i)^{-1} (|F^{NL}_{S,\|}|^2 \cos\theta_S + |F^{NL}_{T,\|}|^2 \cos\theta_T). \tag{22}$$

From the experiment the maximum total transmitted second harmonic intensity occurred at the phase matching angle of $\theta_i^m=37.27°$. At this angle, the total transmitted intensity is ten orders of magnitude stronger than that at the normal incidence. In the experiment we observed total

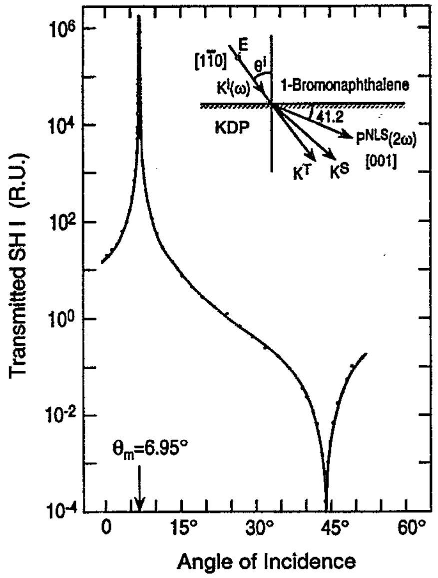

FIG. 6. The total transmitted second harmonic intensity $I_{\text{total}}(2w)=I_S(2w)+I_T(2w)$ as a function of angle of incidence θ_i. The phase matching angle $\theta_i^m=6.95°$ and the dip of $I_{\text{total}}(2w)$ at 44.0°.

transmitted second harmonic intensity as a function of θ_i which varied from $-15°$ to 50°. It is noted that the striking agreement between experimental data points and theoretical curve $I_{\text{total}}(2w)$. In particular, the theoretical prediction by the BP theory of null transmitted second harmonic intensity at $\theta_i=0$ and phase matching angle $\theta_i^m=37.27°$ are confirmed.

C. Phase matched SHG in transmission at oblique angle of incidence and null $I_S(2w)$

The KDP crystal in the experiment has an optic (Z) axis in [001] direction which makes an angle of 41.2° to the crystal entrance surface. The crystal is again immersed in a linear optically denser fluid 1-bromonaphthalene. The fundamental laser beam is polarized in [1$\bar{1}$0] direction which is perpendicular to the plane of incidence as indicated in the inset of Fig. 6. The phase matching angle of KDP is found to be $\theta_m=41.2°$ from the [001] direction. According to the crystallographic cut of the KDP crystal used in the experiment, there exist two possible phase matching directions. The first direction is along the entrance surface of the crystal. This direction is associated with the phase matched SHG at total reflection which has been demonstrated in the previous works.[5–7] The second phase matching direction which is still 41.2° away from optic (Z) axis but on the other side. The angle θ_T corresponding to the phase matching direction is

$$\theta_T=90°-(41.2°+41.2°)=7.6°$$

and the corresponding incidence angle θ_i^m is given by

$$\theta_i^m=\sin^{-1}\left(\frac{n_{2w}^e w(\theta_m)}{n_{\text{liq}}(w)}\sin\theta_T\right),$$

where $n_{2w}^e(\theta_m)=n_w^0=1.4943$. Thus the corresponding θ_i^m is found to be 6.95°. The total transmitted second harmonic intensity $I_{\text{total}}(2w)$ is again given by Eq. (21). The theoretical curve of $I_{\text{total}}(2w)$ is calculated from Eq. (22). The rapid increase of total second harmonic intensity at the phase matching angle θ_i^m is due to an enhancement of nonlinear Fresnel factors $F_{S,\|}^{\text{NL}}$ and $F_{T,\|}^{\text{NL}}$ given by Eqs. (7) and (8). The experimental data dotted points are in striking agreement to the computed theoretical curve using BP theory as shown in Fig. 6.

It is noted that the prominent dip of the curve occurs in the neighborhood of $\theta_i=44.0°$ as expected by the BP theory. The situation of having a dip of total transmitted second harmonic intensity $I_{\text{total}}(2w)$ around the neighborhood of $\theta_i=44.0°$ can be explained as follows. When $\theta_S=(90°-41.2°)=48.8°$ the wave vector $\mathbf{K}_S$ will be parallel to the $\mathbf{P}^{\text{NLS}}$ $(2w)$ which at this situation the angle $\alpha=0°$ and the corresponding incident angle θ_i will be found via Eq. (1) to be $\theta_i=43.73°$ for $\mathbf{K}_S\|\mathbf{P}^{\text{NLS}}$ $(2w)$. According to Eq. (7), the nonlinear Fresnel factor $F_{S,\|}^{\text{NL}}=0$ and this will lead to the inhomogeneous second harmonic intensity $I_S(2w)=0$ at $\theta_i=43.73°$. However, at this angle of incidence, it becomes clear that $I_T(2w)$ is not zero but it has a very small value.

As the angle of incidence θ_i increases to a value of 44.43°, the value corresponding θ_T will be equal to 48.8° and the wave vector $\mathbf{K}^T$ will be parallel to $\mathbf{P}^{\text{NLS}}$ $(2w)$. At this angle of incidence where $\mathbf{K}^T\|\mathbf{P}^{\text{NLS}}$ $(2w)$, the value of α is not zero and $\theta_S\neq\theta_T$. Therefore, from Eq. (8) $F_{T,11}^{\text{NL}}\neq 0$ and consequently $I_T(2w)$ in Eq. (13) will never become zero but rather takes an extremely small value. In conclusion, at the vicinity of $\theta_i=44.0°$ we obtain $I_S(2w)=0$ at $\theta_i=43.73°$ and $I_T(2w)$ becomes very small at $\theta_i=44.42°$, where $\mathbf{K}^T\|\mathbf{P}^{\text{NLS}}$ $(2w)$. Therefore, the value of $I_{\text{total}}(2w)=I_S(2w)+I_T(2w)$ in the neighborhood of $\theta_i=44.0°$ will have a dip as indicated in Fig. 6 and it agrees well to the theoretical curve given by the BP theory.

In addition, it is worthwhile to point out that this particular situation is also predicted by Dick *et al.*[9] According to Eq. (19), the condition for transmitted second harmonic intensity becomes zero is $\alpha+\theta_S+\theta_T=0,\pi$. Since at $\theta_S=48.8°$ or $\theta_i=43.73°$ where $\mathbf{K}^T\|\mathbf{P}^{\text{NLS}}$ $(2w)$ the value of $\alpha=0°$. Therefore, at this angle we have $\theta_S=\theta_R$. Furthermore, at $\theta_i=43.73°$ the reflected angle θ_R is found, via Eq. (1), to be $\theta_R=48.8°$. Under this condition it is found that $\mathbf{K}^{-R}$ vector shown in Fig. 5 is along the direction of $\mathbf{P}^{\text{NLS}}$ $(2w)$. Therefore, the inhomogeneous second harmonic intensity $I_S(2w)=0$. The physical interpretation is that the $\mathbf{P}^{\text{NLS}}$ $(2w)$ cannot radiate into the direction $\mathbf{K}^{-R}$ in the medium which would otherwise lead to the transmitted harmonic wave. This situation is analogous to the situation of nonlinear Brewster angle θ^{NB} condition where $\mathbf{K}^{-T}\|\mathbf{P}^{\text{NLS}}$ $(2w)$ and $\mathbf{P}^{\text{NLS}}$ $(2w)$ cannot radiate in the medium in the direction of $\mathbf{K}^{-T}$ which would otherwise lead to the reflected second harmonic wave.[1]

V. CONCLUSION

The second harmonic generation (SHG) was performed in transmission with various crystallographic cuts of KDP crystals. The investigation of SHG in transmission is investigated in the range of normal incidence to critical angle of incidence. It is for the first time that the null transmitted inhomogeneous and homogeneous second harmonic intensities at normal incidence was observed and demonstrated in good agreement to the prediction of the Bloembergen and Pershan theory[1] and to the prediction of Dick *et al.*[9]

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