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INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS

LAMP SERIES REPORT

(Laser, Atomic and Molecular Physics)

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THEORETICAL PREDICTION OF NONLINEAR BREWSTER ANGLE IN ADP

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Preface

The ICTP-LAMP internal reports consist of manuscripts relevant to seminars and discussions held at ICTP in the field of Laser, Atomic and Molecular Physics (LAMP).

These reports aim at informing LAMP researchers on the activity carried out at ICTP in their field of interest, with the specific purpose of stimulating scientific contacts and collaboration of physicists from Third World Countries.

ABSTRACT

The intensity of reflected second harmonic light from the uniaxial Ammonium di Hydrogen Phosphate, ($NH_4H_2PO_4$)–ADP crystal immersed in the optically denser liquid 1–bromonaphthalene has been theoretically investigated as a function of the incident angle of the fundamental beam of a Nd: YAG laser. The ADP crystal was assumed to be cut with its optics axis [001] inclining at angle $\theta = 42.05°$ to the incident surface. The incident fundamental beam has its polarization along $[1\bar{1}0]$ direction which is normal to the plane of incidence. The intensity of second harmonic light is calculated with the theory of Bloembergen and Pershan. A Nonlinear Brewster Angle $\theta_i^{N.B.}$ of 43.32° is predicted.

Introduction

The behavior of a light wave at the boundary of nonlinear optical medium has been investigated theoretically by Bloembergen and Pershan (BP) [1]. Many features of the BP theory have been verified experimentally in various situations [1-7]. The angular and polarization dependence of the reflected second harmonic intensity was studied by Chang and Ducuing [3]. The Nonlinear Brewster Angle of GaAs was experimentally demonstrated by Chang and Bloembergen [4]. Although Chang and Bloembergen [4] reported of the observation of a Nonlinear Brewster Angle for second harmonic generation, there was always residual second harmonic signal present even at this angle. This was due to the fact that their nonlinear sample GaAs has a complex dielectric constant. The situation was most unfavorable at the ruby laser wavelength. Lee and Bhanthumnavin [7] reported the first experimental verification of a Nonlinear Brewster Angle in second harmonic generation in a transparent dielectric, KDP. In this case all dielectric constants involved are real, and thus the second harmonic signal vanishes exactly at the Nonlinear Brewster Angle.

In this paper, we calculate the conditions for the observation of the Nonlinear Brewster Angle in ADP crystal immersed in the optically denser liquid 1-bromonaphthalene. This same arrangement of the ADP crystal will also permit to observe

phase matched SHG in reflection which occurs at total internal reflection of the incident fundamental beam. We also discuss briefly possible applications of the Nonlinear Brewster Angle's condition to yield the ratio of the two components of the nonlinear polarization $P^{NLS}(2\omega)$ parallel and perpendicular to the incident surface of the crystal. This suggests a zero method as a tool for investigation of nonlinear susceptibility tensors of a crystal. The method is similar to the work of Heinz et al. [8] and B. Dick et. al. [9]

Theory

The geometry of wave vectors of the fundamental and second harmonic light wave at the boundary between 1-bromonaphthalene and unaxial ADP crystal is depicted in Fig. I. The angle θ_R, θ_S and θ_T of the reflected, homogeneous and transmitted wave respectively are given by

$$\eta_{liq}(\omega)\sin\theta_i = \eta_{liq}(2\omega)\sin\theta_R = \eta(\omega)\sin\theta_S = \eta(2\omega)\sin\theta_T \qquad (1)$$

The refractive indices without subscripts refers to the ADP crystal.

The components of nonlinear source polarization P^{NLS} along the cubic axes of the nonlinear ADP crystal are given in terms of the fundamental field components at each point inside the crystal by

$$P_Z^{NLS}(2\omega) = \chi_{36}^{NL} E_x^T(\omega) E_y^T(\omega) \qquad (2)$$

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

If the incident fundamental field is polarized perpendicular to the plane of incidence and along the $[1\,\bar{1}\,0]$ direction ADP (Fig. II) the $P^{NLS}(2\omega)$ will be along [0 0 1] direction and lies in the plane of incidence. Equation (2) can be expressed in terms of the amplitude E_0 of the incident fundamental wave by

$$P_Z^{NLS}(2\omega) = \chi_{36}^{NL}\,\eta\left(F_T^L E_0\right)^2 \qquad (3)$$

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

$$F_T^L = \frac{2\sin\theta_S\cos\theta_i}{\sin\left(\theta_i + \theta_S\right)} \qquad (4)$$

The nonlinear polarization P^{NLS} is the source of harmonic waves. The electric field amplitude of the reflected harmonic wave is related to P^{NLS} by

$$E_R(2\omega) = 4\pi P^{NLS} F_R^{NL} \qquad (5)$$

where F_R^{NL} is the nonlinear Fresnel's factor for the reflected harmonic wave. When P^{NLS} lies in the plane of reflection, F_R^{NL} is given by

$$F^{NL}_{R,II} = \frac{\sin\theta_s \sin^2\theta_T \sin(\alpha + \theta_T + \theta_s)}{\varepsilon_R(2\omega)\sin\theta_R \sin(\theta_T + \theta_R)\cos(\theta_T - \theta_R)\sin(\theta_T + \theta_R)} \qquad (6)$$

where $\sqrt{\varepsilon_R(2\omega)} = \eta_{liq}(2\omega)$

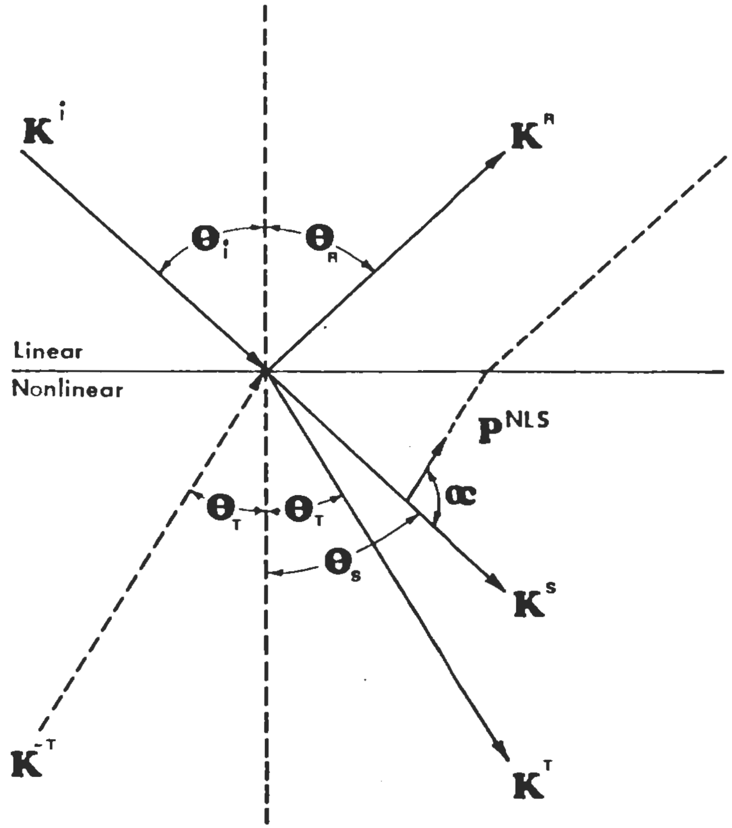

Fig. I Geometry of wave vectors of the fundamental and second harmonic light waves used to illustrate the existence of Nonlinear Brewster Angle

The time averaged second harmonic power carried by the harmonic beams is given by the real part of the poynting vector multiplied by the cross-sectional area A of the various harmonic beams. The power of the reflected harmonics beam is

$$I_R(2\omega) = (C/8\pi)\sqrt{\varepsilon_R(2\omega)}\,|E_R(2\omega)|^2 A \qquad (7)$$

A is the cross-sectional area of the beam and given by

$$A = (dd'\cos\theta_R)/\cos\theta_i \qquad (8)$$

where dd' is the rectangular slit which defines the size of the incident laser beam.
By substitution relevant expressions into (7), we finally obtain

$$I_R(2\omega) = (C/8\pi)\sqrt{\varepsilon_R}\,|E_0|^4\, dd\left(4\pi\chi^{NL}_{36}\right)^2 \eta^2 |F^L|^4 |F^{NL}_R|^2 \cos\theta_R\left(\cos\theta_i\right)^{-1} \qquad (9)$$

By considering factors I_R, dd', χ^{NL}_{36} of ADP, E_0 and η^2 are constants and small variation of $\cos\theta_R$ and $\cos\theta_i$ equation (9) can be written as

$$I_R(2\omega) \;\alpha\; |F^L|^4 |F^{NL}_R|^2 \qquad (10)$$

From equation (5) and (6) it is clear that $I_R(2\omega)$ is zero when

$$\theta_T + \theta_S + \alpha = 0, \pi \qquad (11)$$

This is the Brewster condition for second harmonic generation in reflection. Under this condition P^{NLS}_{II} is parallel to the direction of propagation of the reflected harmonic wave in the nonlinear medium and hence cannot radiate. This nonradiate wave upon refraction back into the linear medium would otherwise give rise to the reflected by in this direction θ_R as shown in Fig. I.

Theoretical Calculation of Nonlinear Brewster Angle in ADP

We consider a unaxial ADP crystal cut with orientation as shown in the inset of Fig. II. The crystal is immersed in an (optically denser) fluid 1-bromonaphthalene which has larger indices of reflection than ADP at both ω and 2ω. This fluid is transparent in the range 0.4-1.6 μm. The index of refraction of the fluid are $\eta_{liq}(\omega) = 1.6260$ and $\eta_{liq}(2\omega) = 1.6701$. The calculation is based on the incident fundamental beam (ω) of Nd: YAG laser, which has $\lambda = 1.06$ μm. The incident beam is polarized in $[1\,\bar{1}\,0]$ direction making incident angle θ^i with respect to the face normal of ADP surface. The nonlinear polarization $P^{NLS}(2\omega)$ is then created in the [0 0 1] direction.

The indices of refraction of ADP obtained from Zernike [11], at are $\lambda = 1.06$ μm, are

$$\eta_o(\omega) = 1.5071, \qquad \eta_e(\omega) = 1.4625$$
$$\eta_o(2\omega) = 1.5284, \qquad \eta_e(2\omega) = 1.4832$$

We have used equation (10) for computation and SHI is given in relative units. In computing the curve, we have to include the angular variation of the refractive index $\eta^e_{2\omega}(\theta)$ inside the ADP crystal, where $\eta^e_{2\omega}(0)$ is given by

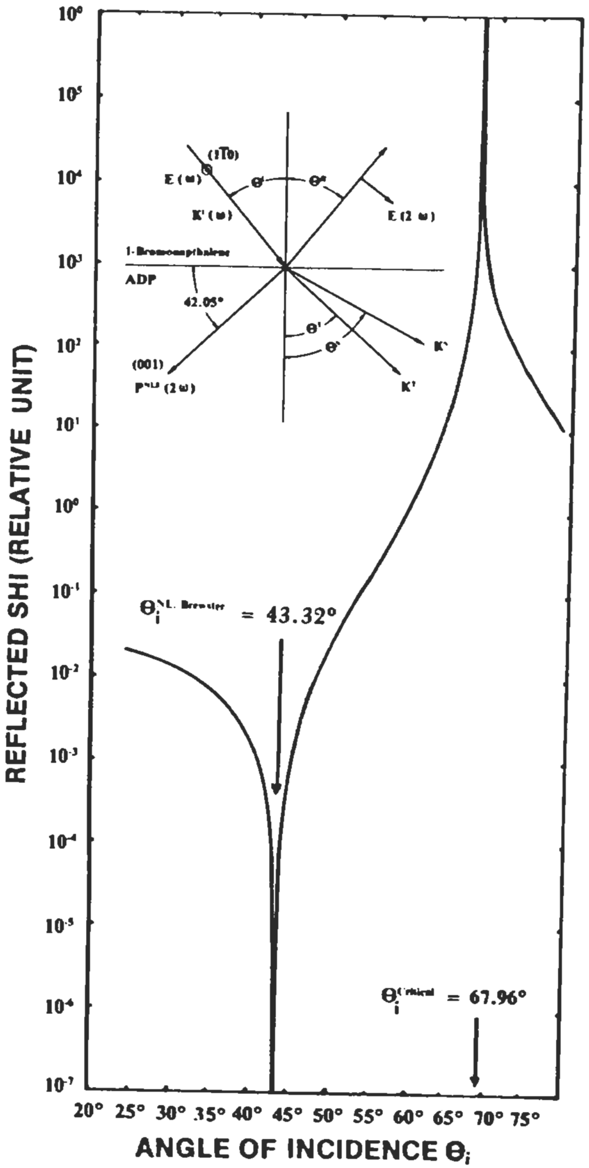

Fig. II The reflected harmonic intensity from the ADP crystal as the function of the angle of incidence. The nonlinear Brewster angle in the case in $\theta^{NB} = 43.32°$. The inset shows the crystal orientation and wave polarization.

$$\frac{1}{\left[\eta^{e}_{2\omega}(\theta)\right]^2} = \frac{1}{\left(\eta^{0}_{2\omega}\right)^2}\cos^2\theta + \frac{1}{\left[\eta^{e}_{2\omega}(\pi/2)\right]^2}\sin^2\theta \tag{12}$$

The pronounced dip at $\theta_i = 43.32^{\circ}$ is due to the Nonlinear Brewster Angle effect. We therefore refer to this angle as Nonlinear Brewster Angle. From the inset in Fig. II, one has $\alpha + \theta_s$, the angle between the nonlinear polarization and the face normal direction of the crystal is equal to -47.95°. Therefore from equation (11), $\theta_T = 47.95^{\circ}$. This together with equation (12), gives $\eta^{e}_{2\omega}(\theta_T = 47.95^{\circ}) = 1.5024$. Thus the Nonlinear Brewster Angle can be determined with the generalized form of Snell's Law, equation (1)

$$\theta_i^{NB} = \sin^{-1}\left(\frac{\eta^{e}_{2\omega}\,\theta_T}{\eta_{liq}(\omega)}\sin\theta_T\right) = 43.32^{\circ}$$

The nonlinear Brewster angle will occur for a particular ratio of the components of the nonlinear polarization perpendicular ($P_{\perp}$) and parallel ($P_{\parallel}$) to the plane of incidence. These in turn will depend on the orientation of the crystallographic axes of the nonlinear optical material with respect to the place of incidence. The situation that we propose for an experiment on ADP is shown in Figure III. In this particular situation the crystallographic axes system (x, y, z) and the surface axes system (ξ, η, ζ) are related through:

$$\begin{pmatrix}\xi\\ \eta\\ \zeta\end{pmatrix} = \frac{1}{-\sqrt{2}}\begin{pmatrix}\sin\varphi & \sin\varphi & -\sqrt{2}\cos\varphi\\ 1 & -1 & 0\\ \cos\varphi & \cos\varphi & -\sqrt{2}\sin\varphi\end{pmatrix}\begin{pmatrix}x\\ y\\ z\end{pmatrix} \tag{13}$$

Hence the component $P_{\parallel}^{NL}$ and $P_{\perp}^{NL}$ are

$$P_{\parallel}^{NL} = P_{\xi} = \frac{1}{\sqrt{2}}(P_x + P_y)\sin\varphi - \sqrt{2}\cos\varphi\ P_z,$$

$$P_{\perp}^{NL} = P_{\xi} = -\frac{1}{\sqrt{2}}(P_x + P_y)\cos\varphi - \sqrt{2}\sin\varphi\ P_z, \tag{14}$$

and their ratio depends on the relative magnitude of P_z and the sum of $P_x + P_y$. Depending on the choice of the polarization of the fundamental beam, P_z and $(P_x + P_y)$ will result from different components of the nonlinear susceptibility tensor, and a measurement of the nonlinear Brewster angle will yield the relative magnitude of these tenser components. Hence the nonlinear Brewster angle could be applicable as a zero-method for the precise measurement of nonlinear optical material constants.

In the case of ADP, cut in the orientation show in Figure III and excited with S-polarized fundamental light, the nonlinear polarization will be along the optic axis for all angle of incidence:

$$P_z = 2d_{36}E_xE_y$$

$$P_x = P_y = 0 \tag{15}$$

Hence the ratio $P_{\perp}/P_{\parallel} = \tan\varphi$ is entirely determined by the geometry of the crystal cut.

The condition for the nonlinear Brewster angle can be written in the form [9]:

$$I^{R}_{2\omega} = \text{const}\left|P_{\parallel}^{NL}\cos\theta_T + P_{\perp}^{NL}\sin\theta_T\right|^2 = 0 \tag{16}$$

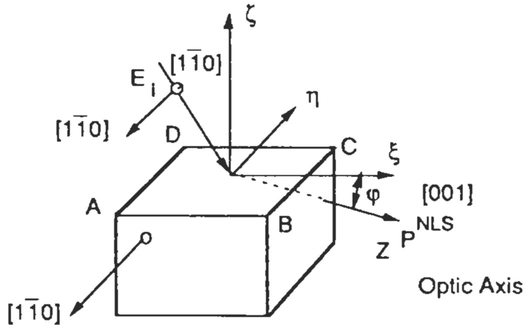

Fig. III Geometry indicating the crystallographic axes (X, Y, Z) and the surface axes (ξ,η,ζ)

Where θ_T is the angle of the transmitted SHG wave inside the crystal as defined in Figure I. At the nonlinear Brewster angle,

$P_{\perp}/P_{\parallel} = \cot\theta_T = \tan\psi$

leading to $\theta_T = 47.95°$.

Conclusion

With the specific transparent ADP crystal cut as indicted in the paper, the Nonlinear Brewster Angle $\theta_i^{NB} = 43.32°$ is theoretically demonstrated. The zero method technique for which the SHG signal vanishes, is briefly analyzed in order to demonstrate the usefulness of the Nonlinear Brewster Angle θ_i^{NB} condition for determining the nonlinear polarization tensor. Furthermore, with this specific ADP crystal cut one can obtain phase matching condition of the reflected second harmonic intensity $I^R(2\omega)$ at total reflection of the fundamental beam.

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