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RELATIVE SIGN AND ABSOLUTE MAGNITUDE OF $\chi^{(2)}$ COEFFICIENTS OF KTP BY SHG MEASUREMENTS

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Abstract

We determine the absolute magnitude of the $\chi^{(2)}$ coefficients of KTP using types I and II phase-matching SHG efficiency measurements : $|\chi_{15}$ (0.532 µm)$|$ = 1.4 ± 0.07 pm/V, $|\chi_{24}$ (0.532 µm)$|$ = 2.65 ± 0.13 pm/V and $|\chi_{33}$ (0.532 µm)$|$ = 10.7 ± 0.5 pm/V. We are the first to establish that these coefficients have the same sign. We compare the obtained magnitudes with the main works previously published.

1. Introduction

KTP is studied and used for its optical quadratic non-linearity since 1976 [1] : the main applications are Second Harmonic Generation (SHG), Optical Parametric Oscillation (OPO) and Optical Parametric Amplification (OPA) in the bulk and more recently in waveguides. The different optical devices including KTP are more and more complicated and efficient. Paradoxally, it exists a big disparity in the different measurements of phase-matching and quadratic non-linear properties of KTP.

The phase-matching directions are directly measured on parallelepipedic or spherical crystals or are more often calculated from Sellmeier's equations previously determined. The coefficients of the second order electric susceptibility tensor $\chi^{(2)}$ are measured by Maker fringe technic or with phase-matched interactions.

An important conclusion of a recent work made by Vanherzeele and Bierlein on KTP [2] using Maker fringe technic is that the measured $\chi^{(2)}$ coefficients are the same whatever the origin of crystals : hydrothermal process (Du Pont) and flux processes (Philips, Ferroxcube, Cristal Laser and Du Pont). The disparity of results between authors is then due to the measurements.

KTP belongs to the crystal class mm2 at a temperature less than its ferroelectric phase transition temperature, equal to 933°C [3,4] ; five coefficients of the second order electric susceptibility tensor $\chi^{(2)}$ ($2\omega = \omega + \omega$) are then independent in the optical frame : χ_{15}, χ_{24}, χ_{31}, χ_{32} and χ_{33}. ω and 2ω are respectively the fundamental and harmonic pulsations.

We determine the absolute magnitude of χ_{15}, χ_{24} and χ_{33} using types I and II phase-matching SHG.

We compare our results with the values obtained by other authors using different methods [1,2,5-12]. Using the sphere method, we are the first to determine the relative signs of χ_{15}, χ_{24} and χ_{33}.

2. Determination of χ_{15} and χ_{24}

In previous works [10,11], we had measured the relative efficiences of two uncoated 1mm - thick strips of KTP cut perpandicular to type II SHG (1.32 µm → 0.66 µm) phase-matching directions contained in the principal planes x-z and y-z. From these measurements, we determine the following ratio :

$$\frac{|\chi_{24}\ (0.66\ \mu m)|}{|\chi_{15}\ (0.66\ \mu m)|} = 1.9 \pm 0.1 \qquad (1)$$

In the present work, we choose type II phase-matched SHG (1.064 µm → 0.532 µm) for the determination of the absolute magnitudes and relative signs of χ_{15} and χ_{24}. We use two uncoated strips of KTP, produced by Cristal Laser, cut perpandicular to the phase-matching directions in the planes x-y and y-z : 0.891 mm thick along ($\theta = 90°$, $\phi = 23.5°$) and 0.888 mm thick along ($\theta = 68.3°$, $\phi = 90°$). The fundamental beam is emitted by a Microcontrôle YAG 904 with the following characteristics : 1W, cw, TEM_{00}, longitudinal multimode, 1-time diffraction-limited. This beam is focused in KTP with an uncoated lens with a focal length l = 10 cm and has a waist *radius* W_o equal to 37 ± 0.7 µm inside the crystal. The CVI coated lens placed after the crystal has a focal length equal to 10 cm. The generated harmonic beam is separated from the non-converted fundamental one by a CVI coated prism. The harmonic power is then detected by a Stanford Research SR530 double phase lock in amplifier and an Hamamatsu S2281-01 photodiode. The fundamental power is controled by an OPHIR 30A-P-CAL powermeter.

In these experimental conditions, we measure the following efficiencies :

$$\eta^{II}_{x\text{-}y} = \frac{P^{II}_{x\text{-}y}\,(0.532\ \mu m)}{P^{\omega}_{o}\,(1.064\ \mu m)} = 8.19 \pm 0.29\ 10^{-6} \quad (2)$$

$$\eta^{II}_{y\text{-}z} = \frac{P^{II}_{y\text{-}z}\,(0.532\ \mu m)}{P^{\omega}_{o}\,(1.064\ \mu m)} = 1.92 \pm 0.07\ 10^{-6} \quad (3)$$

From these low efficiencies, it is possible to determine the absolute value of the effective coefficients $|d_{eff}|$ within the undepleted pump assumption, *i.e.* :

$$|d_{eff}| = \left(\frac{\eta}{A}\right)^{1/2} \quad (4)$$

For a longitudinal multimode fundamental beam, A is expressed by :

$$A = \frac{1.9.10^{-8}\ T_3^{2\omega}\ T_1^{\omega}\ T_2^{\omega}}{\lambda^2\ \cos^4\rho^{2\omega}\ n_3^{2\omega}\ n_1^{\omega}\ n_2^{\omega}}\ \frac{P_o^{\omega}}{W_o^2}\ L^2\ G(\rho,L,W_o) \quad (5)$$

$\lambda(\mu m)$ is the fundamental wavelength. ω is the corresponding circular frequency and 2ω its harmonic.

$P^{\omega}_{o}(W)$ is the incident fundamental power

L(m) is the crystal length.

n_1^{ω}, n_2^{ω} and $n_3^{2\omega}$ are the refractive indices of the three interacting waves in the considered direction. T_1^{ω} T_2^{ω} and $T_3^{2\omega}$ are the corresponding transmission coefficients.

$\rho^{2\omega}$ is the double refraction angle of the harmonic wave.

W_o (m) is the beam *radius*.

$G(\rho,L,W_o)$ is the walk-off attenuation which depends on L,W_o, the type of interaction and on the walk-off angle ρ. G can be calculated assuming the parallel beam limit because the crystal length L (≈ 1mm) is less than the third of the Rayleigh length $Z_0/3$ ($= \pi n^{\omega} W_o^2 / 3\lambda \approx$ 2.5 mm) in our experiments. The calculation of G without approximation leads to a difference of 0.5% [13].

We give in table 1 the previous parameters for the two studied phase-matching directions calculated from the refractive indices given by Kato [7].

According to (2), (3), (4), (5) and table 1, we find :

$$|d_{eff}{}^{II}_{x\text{-}y}\,(1.064\ \mu m \rightarrow 0.532\ \mu m)| = 2.43 \pm 0.12\ pm/V \quad (6)$$

$$|d_{eff}{}^{II}_{y\text{-}z}\,(1.064\ \mu m \rightarrow 0.532\ \mu m)| = 1.29 \pm 0.07\ pm/V \quad (7)$$

d_{eff} is the tensorial contraction of the field tensor $F^{(2)}$, equal to the tensorial product of the three interacting unit electric field vectors, and the unknown second order electric susceptibility tensor $\chi^{(2)}$ [14], *i.e.* for the crystal class mm2 :

$$d_{eff}(\omega+\omega=2\omega) = F_{15}(\omega,2\omega)\,\chi_{15}(2\omega) + F_{24}(\omega,2\omega)\,\chi_{24}(2\omega) + F_{31}(\omega,2\omega)\,\chi_{31}(2\omega) + F_{32}(\omega,2\omega)\,\chi_{32}(2\omega) + F_{33}(\omega,2\omega)\,\chi_{33}(2\omega) \quad (8)$$

The field factors $F_{ij}(\theta,\phi)$ are expressed by :

$$\begin{aligned}
F_{15}(\theta,\phi) &= e^{2\omega}_{x3}(\theta,\phi)\,(e^{\omega}_{x1}(\theta,\phi)\,e^{\omega}_{z2}(\theta,\phi) + e^{\omega}_{z1}(\theta,\phi)\,e^{\omega}_{x2}(\theta,\phi))\\
F_{24}(\theta,\phi) &= e^{2\omega}_{y3}(\theta,\phi)\,(e^{\omega}_{y1}(\theta,\phi)\,e^{\omega}_{z2}(\theta,\phi) + e^{\omega}_{z1}(\theta,\phi)\,e^{\omega}_{y2}(\theta,\phi)\\
F_{31}(\theta,\phi) &= e^{2\omega}_{z3}(\theta,\phi)\,e^{\omega}_{x1}(\theta,\phi)\,e^{\omega}_{x2}(\theta,\phi)\\
F_{32}(\theta,\phi) &= e^{2\omega}_{z3}(\theta,\phi)\,e^{\omega}_{y1}(\theta,\phi)\,e^{\omega}_{y2}(\theta,\phi)\\
F_{33}(\theta,\phi) &= e^{2\omega}_{z3}(\theta,\phi)\,e^{\omega}_{z1}(\theta,\phi)\,e^{\omega}_{z2}(\theta,\phi)
\end{aligned} \quad (9)$$

θ and ϕ are the spherical coordinates of the phase-matching direction in the optical frame.

(x,y,z) refers to the optical frame where z is the binary axis.

The indices 1 and 2 are relative to the fundamental waves and 3 to the harmonic wave. $e^{\omega}_{1}(\theta,\phi)$ and $e^{\omega}_{2}(\theta,\phi)$ correspond to different eigenmodes for type II.

The electric field components (e_x, e_y, e_z) of each wave are calculated from the derivation of the equation of propagation projected on the optical frame [14].

	Type II phase-matched SHG (1.064 µm → 0.532 µm)		Type I Phase-matched SHG (1.064µm→0.532 µm)
W_o = 37 µm Beam waist	θ = 90° ϕ = 23.48°	θ = 68.27° ϕ = 90°	θ = 45.5° ϕ = 37.6°
L Crystal length	891 µm	888 µm	880 µm
$n_3^{2\omega} . n_1^{\omega} . n_2^{\omega}$	5.714	5.6275	5.692
$T = T_3^{2\omega} . T_1^{\omega} . T_2^{\omega}$ Total transmission	0.7789	0.7822	0.7799
$\rho^{2\omega}$ Harmonic double refraction angle	0.27°	0°	0.492°
ρ Walk-off angle	0.27°	1.83°	2.27°
$G(\rho,L,W_o)$ Walk-off attenuation	0.9952	0.8095	0.8752
F_{15}	0.1789	0.9196	0.4794
F_{24}	0.8211	0	-0.5116
F_{31}	0	0	0.01482
F_{32}	0	0	0.01322
F_{33}	0	0	0.02360

Table 1 : Calculated characteristics of the three phase-matching directions used for the determination of χ_{15}, χ_{24} and χ_{33} from conversion efficiency measurements. The calculations are made from the refractive indices given by Kato [7]

We give in table 1 the field factors of the two studied phase-matching directions calculated from the refractive indices given by Kato [7].

The absolute magnitude of χ_{15} at 0.532 µm can be determined from $d_{eff\,y\text{-}z}^{II}$(1.064 µm → 0.532 µm), *i.e.* according to (6), (8) and table 1 :

$$|\chi_{15}(0.532\ \mu m)| = 1.4 \pm 0.07\ pm/V \quad (10)$$

χ_{24} can be deduced from $|\chi_{15}|$ and $|d_{eff_{x-y}}^{II}|$ which is a linear combination of these two coefficients. According to (7), (8), (10), table 1 and the relative sign between χ_{15} and χ_{24}, the two possible values are :

- for different signs :

$$|\chi_{24}(0.532\ \mu m)| = 3.26 \pm 0.16\ pm/V$$

$$i.e\quad \frac{|\chi_{24}(0.532\ \mu m)|}{|\chi_{15}(0.532\ \mu m)|} = 2.3 \pm 0.03 \qquad (11)$$

- for same signs :

$$|\chi_{24}(0.532\ \mu m)| = 2.65 \pm 0.13\ pm/V$$

$$i.e\quad \frac{|\chi_{24}(0.532\ \mu m)|}{|\chi_{15}(0.532\ \mu m)|} = 1.89 \pm 0.03 \qquad (12)$$

The *ratio* (12) is identical to the *ratio* (1), determined at 0.66 µm assuming Miller's rule.

Thus, we can conclude that χ_{15} and χ_{24} have the same signs and that (12) is valid. This sign is taken positive by convention.

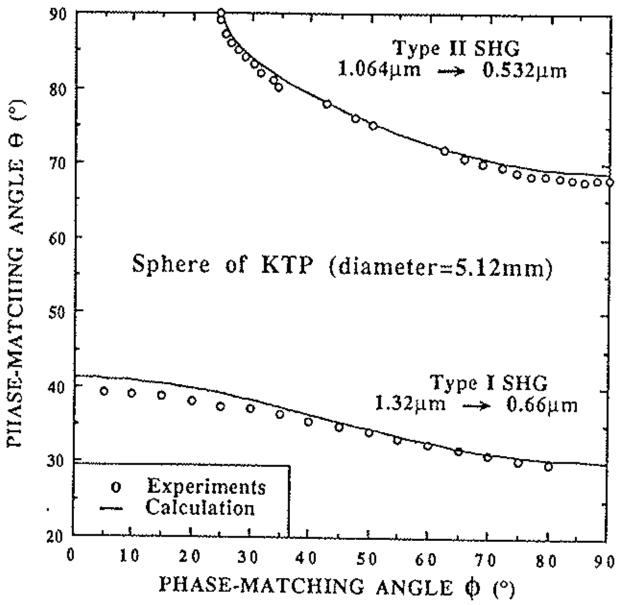

Figure 1 : Spherical coordinates (θ,ϕ) of phase-matched type I SHG (1.32µm→0.66µm) and type II SHG (1.064µm→0.532µm) directions measured in a sphere of KTP

This result is confirmed by an experiment using a KTP crystal cut in a sphere : the conversion efficiencies for several type II phase-matching directions of SHG (1.064 µm → 0.532 µm) contained between the y-z and x-y planes are measured. The interest of such experiments has been demonstrated from 1987 by Velsko [15,16] on spheroidal crystals and from 1989 by us [17] on spherical crystals. We give in figure 1 the spherical coordinates (θ,ϕ) of the phase-matching directions of type II SHG (1.064 µm → 0.532 µm) measured on a sphere of KTP. The agreement is good between the experiment and the calculation made from Sellmeier's equations given by Kato [7]. The associated measured efficiencies are plotted as a function of ϕ in figure 2(a). The theoretical efficiencies are calculated from equations (4), (5) and (9) with the parameters given in table 1, the value of $|\chi_{15}|$ given by (10) and the two possible values of $|\chi_{24}|$ given by (11) and (12).

It clearly appears that the best agreement corresponds to the case where χ_{15} and χ_{24} have the same signs : the efficiency increases from y-z plane to x-y plane. In the contrary hypothesis, the efficiency would reach a nil value at ϕ of about 55° corresponding to the phase-matching direction where the field factors F_{15} and F_{24} are equal, as it is shown in figure 2(b). The field factors show also that type II SHG efficiency in KTP depends essentially on χ_{15} and χ_{24} because the *ratii* between F_{31}, F_{32}, F_{33} and F_{15} or F_{24} are of about several hundred. Actually, F_{31}, F_{32} and F_{33} are biaxial field factors which are all the smaller since n_x approaches n_y, that is to say since the biaxial crystal tends to an uniaxial one [14]. It is the case for KTP where the *ratio* $(n_x - n_y) / (n_z - n_x)$ and $(n_x - n_y) / (n_z - n_y)$ are of about 0.1.

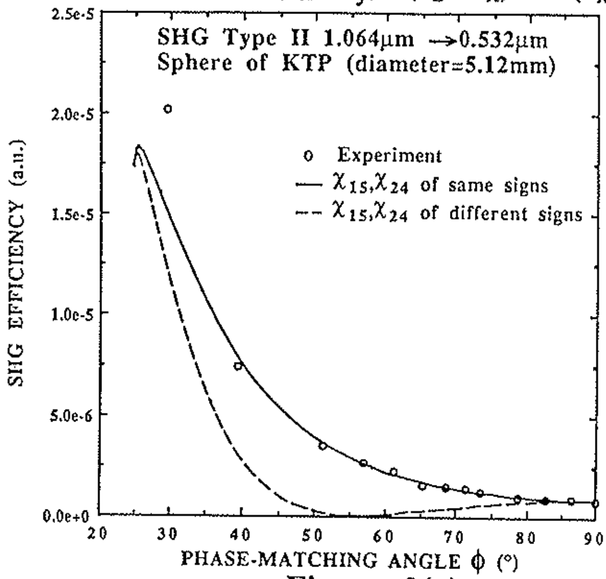

Figure 2(a)
Type II SHG (1,064 μm → 0.532 μm) efficiency measured in a sphere of KTP as a function of the phase-matching direction. The angle θ corresponding to the angle φ is given in figure 1

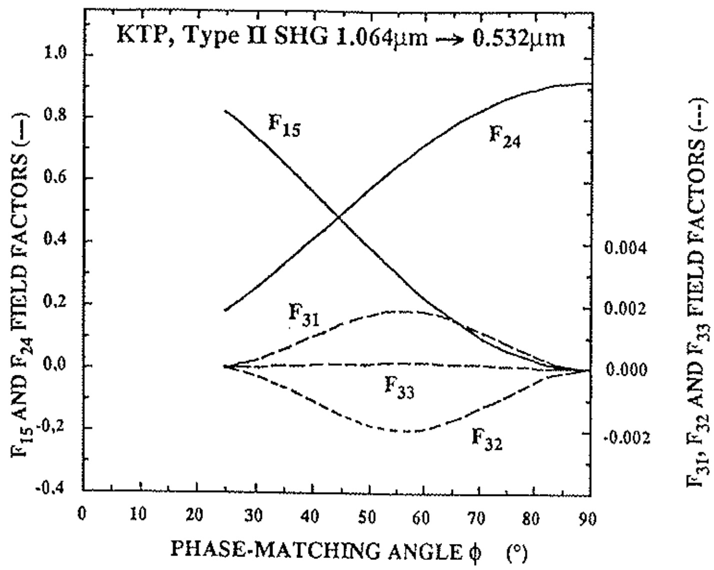

Figure 2(b)
Field factors of type II SHG (1.064 μm → 0.532 μm) in KTP calculated as a function of the phase-matching direction

3. Determination of χ_{33}

The study of the field factors of type I phase-matching SHG in KTP shows that this interaction involves the five independent coefficients χ_{15}, χ_{24}, χ_{31}, χ_{32} and χ_{33} [18]. We give in figure 3(a) the associated field factors for the phase-matched conversion 1.32 μm → 0.66 μm, calculated with the refractive indices given by Kato [7] from equations (9), where $\mathbf{e}_1^{\omega}(\theta,\phi)$ and $\mathbf{e}_2^{\omega}(\theta,\phi)$ are identical by definition of type I. The "quasi-uniaxiality" of KTP leads to the three following relations :

$$F_{15}(\theta,\phi) \approx -F_{24}(\theta,\phi) \quad (13)$$

$$F_{31}(\theta,\phi) \approx F_{32}\left(\theta,\phi+\frac{\pi}{2}\right) \quad (14)$$

$$|F_{15} + F_{24}| \approx (F_{31}, F_{32}, F_{33}) \quad (15)$$

The five $\chi^{(2)}$ coefficients are involved because of (15). Unfortunatelly, they cannot be determined from five type I SHG efficiency measurements made in five different phase-matching directions. Actually, because of relations (13) and (14), the convergence is difficult to reach for the resolution of the system of the five linear equations (8) which would be deduced from these measurements. On the other hand, an accurate determination of χ_{33} is possible from type I measurements if the values of χ_{15} and χ_{24}, well determined by type II phase-matching SHG, are taken into account and if Kleinman symmetry assumption is made, *i.e.* :

$$\chi_{15} = \chi_{31} \qquad \chi_{24} = \chi_{32} \qquad \chi_{33} \quad (16)$$

The assumption of Kleinman symmetry is totally justified for KTP because the electric polarization is of electronic origin and the optical absorption at the concerned wavelengths are very low, less than 1% cm^{-1} at 0.532 µm and less than 0.1% cm^{-1} at 1.064 µm for KTP crystals produced by Cristal Laser. Thus, the gaps between χ_{15} and χ_{31} on one hand and between χ_{24} and χ_{32} on the other hand should be very weak and assuredly contained within the uncertainty of our efficiency measurements, of about ± 3.5%.

Thus, the determination of χ_{33} by type I phase-matched SHG is accurate within Kleinman assumption, all the more so since the difference between the real effective coefficient d_{eff}^{real} and $d_{eff}^{Kleinman}$ depends on F_{31} and F_{32} which are very low :

$$|d_{eff}^{real} - d_{eff}^{Kleinman}| = F_{31}(\chi_{31} - \chi_{15}) + F_{32}(\chi_{32} - \chi_{24}) \approx 0 \qquad (17)$$

We choose to use the phase-matching direction of maximum efficiency, which is the less sensitive to an angular shift, for the determination of χ_{33}. For the conversion 1.064 µm → 0.532 µm, the spherical coordinates of this direction measured on sphere are θ = 45.5° and ϕ = 37.6°.

The conversion efficiency η_{max}^{I} measured in an uncoated 0.880 mm thick strip of KTP cut perpandicular to this direction is :

$$\eta_{max}^{I}(1.064\ \mu m \rightarrow 0.532\ \mu m) = 1.67 \pm 0.06\ 10^{-7} \qquad (18)$$

According to *formulae* (4), (5) and the parameters of the considered direction given in table 1, the effective coefficient deduced from (18) is :

$$|d_{eff\,max}^{I}(1.064\ \mu m \rightarrow 0.532\ \mu m)| = 0.37 \pm 0.02\ pm/V \qquad (19)$$

The value of χ_{33} depends on the sign of $d_{eff\,max}^{I}$. The two possible values of χ_{33} deduced from (19) are the following, according to *formula* (8), to the field factors given in table 1, to the values of χ_{15} and χ_{24} given by (10) and (12) respectively and according to Kleinman assumption :

$$\chi_{33}(0.532\ \mu m) = +39.5 \pm 2\ pm/V \quad \text{for} \quad d_{eff\,max}^{I} > 0 \qquad (20)$$

$$\chi_{33}(0.532\ \mu m) = +10.7 \pm 0.5\ pm/V \quad \text{for} \quad deff_{max}^{I} < 0 \qquad (21)$$

The first conclusion is that χ_{33} has the same sign as χ_{15} and χ_{24}, which has never been established by previous works.

This result is confirmed by a sphere experiment. We take as example type I phase-matching for the conversion 1.32 µm → 0.66 µm. The spherical coordinates (θ,ϕ) of the measured phase-matching directions are given in figure 1. The associated efficiencies are plotted as a function of ϕ in figure 3(b). We also give in figure 3(b) the two theoretical ϕ angular variations of type I phase-matching SHG (1.32 µm → 0.66 µm) efficiency calculated with the two possible values of χ_{33} at 0.66 µm deduced from (20) and (21) by Miller's rule. The agreement with the experimental *data* is the best in the case of a negative effective coefficient which corresponds to the value of χ_{33} given by (21).

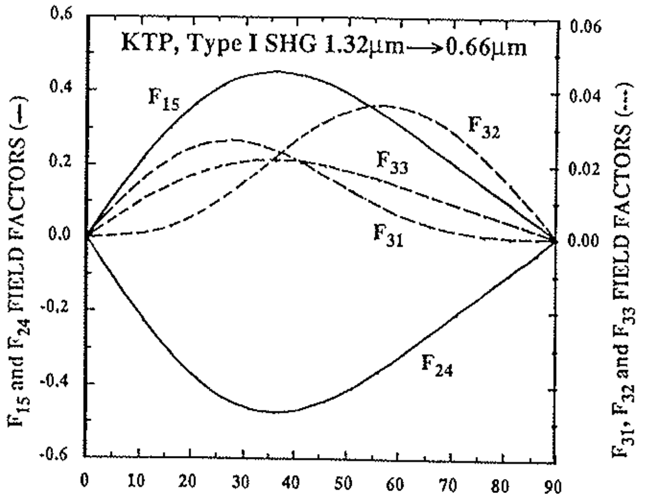

Figure 3(a)
Field factors of
type I SHG (1.32 μm → 0.66 μm)
in KTP, calculated as a function
of the phase-matching direction

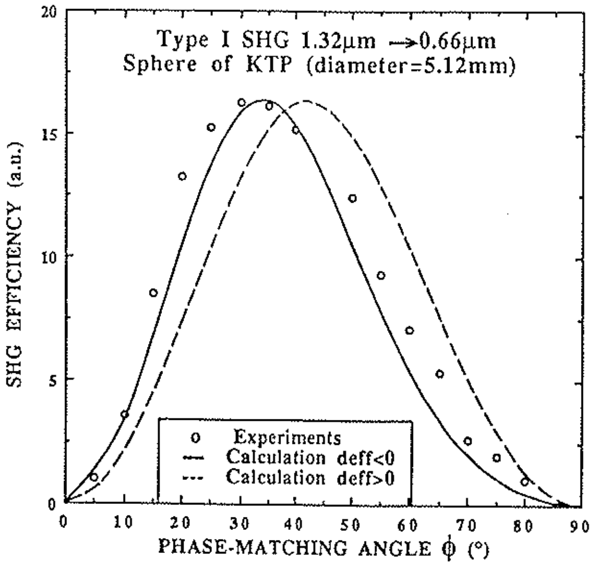

Figure 3(b)
Type I SHG (1.32 μm → 0.66 μm)
efficiency measured in a sphere of KTP
as a function of the phase-matching direction.
The correspondence between θ and ϕ
is given in figure 1

4. Comparison with previous works and conclusion

The main results, including this work, concerning the measurements of the optical quadratic non-linearity of KTP are summarized in table 2. Zumsteg *et al* [1] and Vanherzeele *et al* [2] use Maker fringe method, Nishikawa *et al* [5], Eckardt *et al* [6], Kato [7,9], Zondy [12] and us [10,11] use phase-matched SHG (or OPO) and DeSalvo *et al* [8] use a cascaded $\chi^{(2)} : \chi^{(2)}$ process near SHG phase-matching. The value of χ_{24} at 0.532 μm given by Zondy *et al* is deduced by Miller's rule from a *datum* at 0.65 μm obtained by type II phase-matched SHG efficiency measurement in x-z plane at a fundamental wavelength of 1.3 μm, *i.e.* $|d_{eff_{xz}}^{II}|$ (1.3 μm → 0.65 μm) = 2.10 ± 0.17 pm/V.

Note that the wavelengths of 1.064 μm and 0.88 μm given in table 2 of the publication of Vanherzeele *et al* [2] are the fundamental wavelengths used for their Maker fringe experiments and not the wavelengths of the corresponding $\chi^{(2)}$ coefficients : they are obviously measured at 0.532 μm and 0.44 μm.

The agreement between magnitudes is good for the following results :

- the effective coefficient of type II phase-matched SHG in x-y plane given by Eckardt *et al*, Vanherzeele *et al* and DeSalvo *et al*,
- the effective coefficient of type II phase-matched SHG in y-z plane given by Kato and Vanherzeele *et al*,
- the relative value of the effective coefficients of types I and II given by Vanherzeele *et al*, Nishikawa *et al* and us,
- χ_{33}/χ_{15} obtained by Vanherzeele *et al* and us,
- χ_{24}/χ_{15} obtained by Kato, Vanherzeele *et al* and us,
- χ_{24} given by Zondy *et al* and us,
- χ_{33} given by Zumsteg, Kato and us.

Quadratic optical non-linearity of KTP									
Authors		Zumsteg *et al* 1976 [1]	Nishikawa *et al* 1989 [5]	Eckardt *et al* 1990 [6]	Kato 1991-1992	DeSalvo *et al* 1992 [8]	Vanherzeele *et al* 1992 [2]	Our measurements 1992-1993 [10,11]	Zondy *et al* 1993 [12]
Method		Maker fringe	Type I, Type II phase-matched SHG	Type II phase-matched SHG	Type I, Type II phase-matched SHG, OPO	Cascaded $\chi^{(2)} : \chi^{(2)}$ effects	Maker fringe	Type I, Type II phase-matched SHG	Type II phase-match. SHG
Accuracy		-	-	± 5%	±10%	-	±10%	±5%	±8%
$\chi(0.532\mu m)$ (pmV^{-1})	χ_{15}	$\lvert 6.1 \rvert$*	-	$\lvert 2.6 \rvert$	$\lvert 1.9 \rvert$	-	$\lvert 1.91 \rvert$*	+ 1.4	-
	χ_{24}	$\lvert 7.6 \rvert$*	-	$\lvert 3.3 \rvert$	$\lvert 3.4 \rvert$	-	$\lvert 3.64 \rvert$*	+ 2.65	2.58
	χ_{31}	$\lvert 6.5 \rvert$*	-	-	$\lvert 1.9 \rvert$	-	$\lvert 2.54 \rvert$*	+ 1.4	-
	χ_{32}	$\lvert 5.0 \rvert$*	-	-	$\lvert 3.4 \rvert$	-	$\lvert 4.35 \rvert$*	+ 2.65	-
	χ_{33}	$\lvert 13.7 \rvert$*	-	-	$\lvert 8.1 \rvert$	-	$\lvert 16.9 \rvert$*	+ 10.7	-
χ_{33}/χ_{15}		2.25	-	-	4.26	-	8.85	7.57	-
χ_{24}/χ_{15}		1.24	-	idem Zumsteg	1.80* [7]	-	1.91	1.89 [10]	-
$d_{eff}(pmV^{-1})$ Type II phase-match. SHG 1.064→0.532 $\theta = 90°$		7.32	-	3.2*	idem Eckardt	3.1	3.35	2.43*	-
$d_{eff}(pmV^{-1})$ Type II phase-match. SHG 1.064→0.532 $\phi = 90°$		5.61	4.6*	2.4	1.75	-	1.88	1.295*	-
$d_{eff}(pmV^{-1})$ Type I phase-match. SHG 1.064→0.532	ϕ=37.6	-0.479	-	-	-0.565	-	-0.488	-0.372*	-
	ϕ=45.7	-0.459	-	-	-0.540	-	-0.461	-0.355	-
	ϕ=79.7	-0.121	0.40*	-	-0.141	-	-0.113	-0.0908	-
$\frac{d_{eff}^{II} (\phi=90)}{d_{eff}^{I} (\phi=45.7)}$		12.2	-	-	3.0* [9]	-	4.08	3.65	-
$\frac{d_{eff}^{II} (\phi=90)}{d_{eff}^{I} (\phi=79.7)}$		46.4	11.5	-	idem Nishikawa	-	16.6	14.3	-

Table 2 : Main published results on the quadratic optical nonlinearity of KTP.

*Experimental *data* : d_{eff}* measured by phase-matched method, from which the χ_{ij} and the other d_{eff} are calculated. χ_{ij}* measured by Maker fringe method. from which the different d_{eff} are calculated.

It appears that Kleinman's conditions are not satisfied according to Maker fringe measurements contrary to our hypothesis. However, if we take the *ratio* between χ_{15} and χ_{31} and the *ratio* between χ_{24} and χ_{32} obained by Vanherzeele *et al* for the interpretation of our type I SHG experiments, we find a value of χ_{33} equal to 10 pm/V, instead of 10.7 pm/V within Kleinman assumption. This result confirms the remarks made in §3 concerning the accuracy of type I phase-matching SHG for the determination of χ_{33}.

Among all the previously published works, we are the only to determine the relative sign of χ_{15}, χ_{24} and χ_{33}.

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