An upper limit on all relaxation rates is obtained by setting all energy differences in (D6c) and (D6d) equal to zero. This limit is even realizable in practice, since $8\Psi_d$, converted to units of Oe, using $g=2.0$,[29] has the value

$$8\Psi_d\hbar/(2\mu_B)=18\ 600\ \text{Oe}, \tag{D8}$$

which not only is well above the energy level differences at zero external field, but is even large compared to typical values of the external field in actual experiments.[17,23] In the limit where all energy differences are small compared to (D8), we have

$$\zeta_{nm}^{(2)}=[\langle n|h^2|n\rangle+\langle m|h^2|m\rangle-2\langle n|h|n\rangle\times\langle m|h|m\rangle]/8\Psi_d, \tag{D9a}$$

$$\zeta_{nj}^{(1)}=2|\langle n|h|j\rangle|^2/8\Psi_d. \tag{D9b}$$

The relaxation rates $\zeta_{nm}^{(2)}$ as well as the total diagonal relaxation rates $\sum_j\zeta_{nj}^{(1)}$ are all seen to be bounded from above by the maximum value attainable by the quantity $\langle n|h^2|m\rangle/4\Psi_d$. Since $h^2=(E')^2S_{z'}^2$, this maximum value is $(E')^2/4\Psi_d$, which in units of Oe is about 6 Oe. Except near degeneracies, this value is indeed well below all finite energy differences. Similarly, the energy corrections ϵ_n are smaller than typical energy differences by at least a factor $(E'/8\Psi_d)^2\approx 2\times10^{-4}$. Thus the assumptions leading to (D6a) are justified, provided there are no near degeneracies and provided Dk^2 is small compared to all finite energy differences. The k's of interest will necessarily correspond to distances of the order of a diffusion length, $(D/\zeta)^{1/2}$, where ζ is a typical relaxation rate. Thus Dk^2 is of the order of ζ and is sufficiently small. Finally, the small magnitude of the relaxation rates allows us to ignore near degeneracies: Such degeneracies will modify the relaxation rates at worst only when the corresponding energy difference becomes comparable to 6 Oe, in appropriate units, a very narrow energy range compared to the full range. Nor do such modifications significantly affect the resonance shapes; the resonance shapes will be affected only in a range of at most 6 Oe about the center of the resonance; by contrast, the resonance width will be of the order of $2.24(\zeta\Psi_d)^{1/2}\approx|E'|\sim250$ Oe, even when relaxation is the only decay mechanism.

For external fields well below the value (D8), the average diagonal relaxation rate is given by

$$\zeta=\tfrac{1}{3}\sum_n(\sum_j\zeta_{nj}^{(1)})=(E')^2/6\Psi_d$$
$$=8.2\times10^7\ \text{sec}^{-1}\quad(\hbar\zeta/2\mu_B=4.65\ \text{Oe}). \tag{D10}$$

This is also approximately the average of the off-diagonal relaxation rates. If the expression (D9a) is averaged over all n, m, ignoring the restriction $n\neq m$, the result is precisely (D10).

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Optical Nonlinear Susceptibilities: Accurate Relative Values for Quartz, Ammonium Dihydrogen Phosphate, and Potassium Dihydrogen Phosphate

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A complete theoretical and experimental analysis of the Maker fringes leads to the determination of accurate relative nonlinear optical susceptibilities in SiO_2, $NH_4H_2PO_4$, and KH_2PO_4: $d_{11}^{2\omega}(SiO_2)=(0.77\pm0.04)\ d_{36}^{2\omega}(KH_2PO_4)$; $d_{36}^{2\omega}(NH_4H_2PO_4)=(1.21\pm0.05)d_{36}^{2\omega}(KH_2PO_4)$, for a fundamental wavelength 1.064 μm. Application of this analysis to earlier data leads to corrected values in good agreement with the present study, thereby establishing these materials as accurate and reliable standards for measurement of nonlinear optical susceptibilities.

INTRODUCTION

SINCE it is extremely difficult to make accurate absolute-intensity measurements, the components of the tensor describing optical second-harmonic generation or parametric processes have with few exceptions been determined by relative measurements: the elements of the unknown tensor being compared to those of a well-known "standard" material, such as KH_2PO_4 (KDP). Accurate absolute values can then be easily deduced from relative data, provided the standard has been carefully calibrated.

A survey of all the published measurements[1] indicates, however, a complex and quite paradoxical situation. Most importantly, no one material has been chosen as a common standard. Depending on the spectral range (visible, infrared, ultraviolet) and on the techniques

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[1] R. Bechmann and S. K. Kurtz, *Landolt-Börnstein: Numerical Data and Functional Relastionships, Group III. Crystal and Solid State Physics* (Springer-Verlag, Berlin, 1969), Vol. 2.

used for the determination of the nonlinear coefficients (phase-matching experiment,[2,3] reflection at the boundaries of the nonlinear medium,[4,5] Maker fringe experiment[3]), the reference materials are different. For the visible and ultraviolet region two reference materials are frequently encountered in the literature: quartz and KDP. By contrast, the most accurate and reliable absolute determination of a nonlinear coefficient has been made for $NH_4H_2PO_4$ (ADP).[6,7]

Lack of reliable ratios for the three reference materials, quartz, KDP, and ADP, has led to considerable uncertainty in the conversion of nonlinear coefficients to absolute values. Consequently, the relative calibration of these reference materials is of great importance from the fundamental point of view as well as for applications such as optical parametric oscillators. Indeed, there are several publications on this subject,[8–13] but the different ratios are inconsistent: the discrepancy among the data concerning, for example, the fundamental ratio $d_{36}{}^{2\omega}(\mathrm{ADP})/d_{36}{}^{2\omega}(\mathrm{KDP})$ equals several times the quoted experimental errors.

We report in this paper the precise relative determination of $d_{11}{}^{2\omega}$(quartz), $d_{36}{}^{2\omega}$(KDP), and $d_{36}{}^{2\omega}$(ADP) using a detailed analysis of the Maker-fringe technique[14] which leads to an accuracy greater than 5%. Our results are then compared with the previously published data.

I. THEORY

Owing to the existence in a nonlinear crystal of a polarization $\mathcal{P}_{2\omega}$, which is quadratic in the applied electric field, the second-harmonic beam can, in the parametric approximation, be described as a superposition of two waves.[4,15] One wave, the so-called "bound wave," is induced by $\mathcal{P}_{2\omega}$ acting as a source term in the wave equation. Boundary conditions at the input surface of the crystal give rise to the second, or so-called "free wave," which interferes with the bound wave at the output surface to produce a transmitted second-harmonic beam. If a Gaussian laser beam, with spot radius w and power P_ω, is incident at various angles θ on a nonlinear plane parallel slab of thickness L, the detected second-harmonic power $P_{2\omega}$ is given by [14]

$$P_{2\omega}(\theta)=\frac{512\pi^2}{cw^2}d^2 l_\omega{}^4(\theta)T_{2\omega}(\theta)p^2(\theta)\mathfrak{R}(\theta)\mathfrak{B}(\theta)\frac{P_\omega{}^2}{(n_\omega{}^2-n_{2\omega}{}^2)^2}$$
$$\times\sin^2[(\omega L/c)(n_\omega\cos\theta_\omega{}'-n_{2\omega}\cos\theta_{2\omega}{}')]\equiv I_M(\theta)\sin^2\Psi, \quad (1)$$

where d is the component of the quadratic susceptibility tensor $\mathbf{d}$, and t^ω and $T_{2\omega}$ are the transmission factors for the fundamental and harmonic frequencies. The projection factor $p(\theta)$ depends on the orientation of the slab versus the laser beam and on the form of the tensor $\mathbf{d}$; $\theta_\omega{}'$ and $\theta_{2\omega}{}'$ are the angles between the normal to the slab and the internal wave vectors of the fundamental and harmonic beams, n_ω and $n_{2\omega}$ being the refractive indices at the fundamental and harmonic frequencies, respectively.

The relation between $P_{2\omega}$ and P_ω has been established for a transparent and nonoptically active crystal including two generally omitted effects. One is multiple reflections inside the crystal which introduce the factor $\mathfrak{R}(\theta)$. An additional factor $\mathfrak{B}(\theta)$ takes into account the finite transverse size of the laser beam. The multiple-reflection factor $\mathfrak{R}$ is close to unity as long as the refractive indices are low. The finite-beam factor $\mathfrak{B}(\theta)$ is given by [14]

$$\mathfrak{B}(\theta)=\exp\{-[(L^2/w^2)(\tan\theta_\omega{}'-\tan\theta_{2\omega}{}')^2\cos^2\theta]\} \quad (2)$$

and is non-negligible for thick crystals at large values of θ.

Linear dispersion $(n_\omega-n_{2\omega})$ is responsible for interference phenomena inducing $P_{2\omega}$ to be an oscillating function of θ described by[4]

$$\sin^2\Psi=\sin^2(\omega L/c)(n_\omega\cos\theta_\omega{}'-n_{2\omega}\cos\theta_{2\omega}{}')$$
$$=\sin^2\tfrac{1}{2}\pi[L/l_c(\theta)]. \quad (3)$$

From the structure of the Maker fringes, one can therefore determine the coherence length l_c for normal incidence:

$$l_c=\frac{\pi c}{2\omega|n_\omega-n_{2\omega}|}. \quad (4)$$

With the laser operating under identical conditions, relative nonlinear coefficients can be measured using the Maker fringes by comparing the value of the envelope function at normal incidence $I_M(0)$ for an unknown material with $I_M(0)$ for a reference material. According to Eq. (1) the relationship between $I_M(0)$, l_c, and d is

$$d^2=A\frac{I_M(0)}{l_c{}^2}\left(\frac{(n_\omega+1)^3(n_{2\omega}+1)^3(n_\omega+n_{2\omega})}{n_{2\omega}p^2(0)\mathfrak{R}(0)}\right)$$
$$\equiv A\frac{I_M(0)}{l_c{}^2}\eta, \quad (5)$$

where [14] $\mathfrak{R}(0)\simeq 1+[(n_{2\omega}-1)/(n_{2\omega}+1)]^4$ and A is constant for a given fundamental beam. Since only a few

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significant figures for the refractive indices provide a precise value of η, the accuracy of the method depends chiefly on the precision with which $I_M(0)$ and l_c are determined.

II. EXPERIMENTAL DATA

A repetitively Q-switched Nd^{3+}YAG laser having linear polarization was used as the fundamental beam. Because oscillations on several $4F_{3/2}\rightarrow 4I_{11/2}$ transitions can occur simultaneously,[16] the output spectrum of the laser was analyzed using a monochromator[17,18] and was found to be a single line of 0.6 cm^{-1} width at 1.064 μm.

In order to check the stability of the laser, the second harmonic generated in a fixed quartz sample was recorded. Fluctuations in the fundamental power were thus taken into account for the rotating samples.

Experiments involved several plates of the three materials quartz, ADP, and KDP, as indicated in Table I. Care was taken, during the preparation of every sample, to obtain faces flat to $\frac{1}{2}\lambda$ and parallel within 1′ or less.

A. Determination of Coherence Length l_c

For each sample, the analysis of the positions of the minima (which can be experimentally determined with better accuracy than the maxima) of the Maker fringes for a wide range of θ, leads to a very precise determination of the coherence length l_c.[14] No detectable difference has been found between samples of the same material. The experimental results are listed in Table II and compared with the values calculated from data on the refractive indices.[19–21] In the case of KDP and ADP, it appears there is a good agreement with Zernike's data,[19] but a non-negligible discrepancy, especially for ADP, with the results given by Suvorov and Sonin.[20]

B. Relative Envelope Intensities $I_M(0)$

This part of the measurement is the most important source of errors.

Because we are dealing with relative measurements, there is no need to calibrate the detection system, provided it operates under the same conditions for all the materials. In that respect the linearity of the response at 0.532 μm was checked by attenuating the second-harmonic beam using calibrated filters. It is worth noting that the amplitudes of the Maker fringes are of the same order of magnitude for the three materials and the different components of the detection system were operating at the same sensitivity.

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TABLE I. Crystallographic orientation and thickness of the samples used in this experiment. We indicate two values [(a) and (b)] for the thickness: (a) Corresponds to a direct measurement, and (b) is the value deduced from the analysis of the Maker fringes.

Material	Sample orientation	Thickness (a) (cm)	Thickness (b) (μm)
Quartz	(011)	0.223	2226.8
Quartz	(011)	0.198	1968.2
Quartz	(010)	0.448	4477.5
ADP	(110)	0.550	5507.8
ADP	(110)	0.167	1664.3
KDP	(110)	0.201	2011.6
KDP	(110)	0.215	2154.7

In order to check the consistency of the results, the three ratios

$$\frac{I_M{}^{Q}(0)}{I_M{}^{KDP}(0)},\quad \frac{I_M{}^{Q}(0)}{I_M{}^{ADP}(0)},\quad \frac{I_M{}^{ADP}(0)}{I_M{}^{KDP}(0)}$$

have been determined separately. For a given ratio, Maker fringes corresponding to one sample of each material have been performed for every combination of two samples.

Owing to the adequate flatness and the parallelism of the faces for the samples used, there were no "nonzero minima" corrections.[14] Fluctuations of the quartz reference were taken into account.

Using the amplitudes of the maxima as input data for a least-mean-squares curve-fitting computer program, we determined, according to Eq. (1), the envelope of each set of fringes. A typical example of the fit is given, for each material, in Fig. 1. Previous experiments[14] showed that an optimum fit can be reached by analyzing only a few fringes. We considered, therefore, a relatively small range of variations for θ ($|\theta|\leq 25°$). As a further proof of the consistency of our analysis, theoretical fringes deduced from the computed values of l_c and $I_M(0)$ agree very well with the experimental curves of Fig. 1 as shown for quartz.

Using this procedure the average relative envelope intensities have been found to be

$$I_M{}^{Q}(0)/I_M{}^{ADP}(0)=1.39,\quad \text{rms deviation } 0.04$$
$$I_M{}^{Q}(0)/I_M{}^{KDP}(0)=1.71,\quad \text{rms deviation } 0.06$$
$$I_M{}^{ADP}(0)/I_M{}^{KDP}(0)=1.22,\quad \text{rms deviation } 0.04.$$

TABLE II. Comparison of experimental values of the coherence lengths at 1.064 μm of quartz, KDP, and ADP with the values deduced from published data on the refractive indices.

Material	Experimental value	Calculated value	Reference
Quartz	20.65±0.05	20.636±0.002	21
ADP	10.59±0.02	10.598±0.002	19
		11.56 ±0.1	20
KDP	11.43±0.02	11.417±0.002	19
		11.67 ±0.1	20

TABLE III. Comparison with previous measurements of nonlinear optical coefficients of quartz and ADP relative to KDP. The corrections, prompted by our measurements, of previously published values are given in parentheses. For the Maker-fringe measurements, experimental values of the coherence lengths are also indicated. MF: Maker fringe; PM: phase matching.

Material	Method	λ (μm)	d	l_c	Reference
KDP	MF	1.064	$d_{36}\equiv 1$	11.43±0.02	This experiment
	MF	1.0582	$d_{36}\equiv 1$	11.0	8
	MF	0.6943	$d_{36}\equiv 1$	9.25	8
	MF	0.6943	$d_{36}\equiv 1$	9.8	11
	MF	0.6943	$d_{36}\equiv 1$	9	12
ADP	MF	1.064	$d_{36}=1.21\pm0.05$	10.59±0.02	This experiment
	MF	1.0582	$d_{36}=0.99\pm0.06$	10.5	8
			$(d_{36}=1.18\pm0.06)$		25
	MF	0.6943	$d_{36}=0.93\pm0.06$	8.85	8
			$(d_{36}=1.03\pm0.06)$		25
	MF	0.6943	$d_{36}=1.25\pm0.05$	9.3	11
	MF	0.6943	$d_{36}=1.18\pm15\%$	7.9	12
	PM	1.15	$d_{36}=0.87\pm 9\%$		13
			$(d_{36}=1.03\pm0.16)$		26
Quartz	MF	1.064	$d_{11}=0.77\pm0.04$	20.65±0.05	This experiment
	MF	1.0582	$d_{11}=0.82\pm0.04$	20	9
			$(d_{11}=0.85\pm0.05)$		25

These results indicate random errors less than ±5%. An upper limit of the remaining systematic errors is ±5%, which leads to an accuracy on the relative intensities better than ±10%.

C. Relative Nonlinear Coefficients

In the experimental configuration, the projection factor $p(0)$ equals 1 for the three coefficients d_{11} (quartz), d_{36}(ADP), and d_{36}(KDP). In addition, the refractive indices of the materials are low enough to allow us to take $\mathcal{R}(0)=1$. The error introduced by this approximation[14] is less than 2×10^{-4}.

The final computations involving Eq. (5) lead to the following results:

$$d_{36}(\text{ADP})=(1.57\pm0.07)d_{11}(\text{Q}),$$
$$d_{36}(\text{KDP})=(1.30\pm0.06)d_{11}(\text{Q}),$$
$$d_{36}(\text{ADP})=(1.21\pm0.05)d_{36}(\text{KDP}).$$

A simple way to check the consistency of the quoted values is to deduce $d_{36}(\text{ADP})/d_{36}(\text{KDP})$ from the two first data, which gives

$$d_{36}(\text{ADP})=(1.20\pm0.09)d_{36}(\text{KDP}).$$

III. DISCUSSION

A. Absorption and Optical Activity

To analyze our data and deduce the relative values of the nonlinear susceptibilities, we assumed that quartz, ADP, and KDP are without any optical activity and perfectly transparent for both the 1.064- and 0.532-μm radiations. Let us now consider these assumptions in greater detail.

When there are small absorption coefficients at ω and 2ω, the measured value of the intensity $I_M(0)$ differs from that involved in Eq. (5) by the factor $\mathcal{A}$, which is given by[22]

$$\mathcal{A}=\exp[-(a_\omega+\tfrac{1}{2}a_{2\omega})L], \tag{6}$$

a being the absorption coefficient, as customarily defined. Computations using absorption coefficients reported in the literature[22] show the influence of $\mathcal{A}$ to be negligible in the case of KDP and quartz, because of the small values of L (see Table I). There is for ADP an absorption band with an edge around 1.15 μm,[21,22] but the correction introduced by this absorption remains lower than the error on the determination of $I_M(0)$. No difference has been detected experimentally between the 0.550 and 0.167-cm-thick samples.

Optical activity gives rise to a rotation ρ of the plane of polarization which can be written as a function of the gyration tensor $\mathbf{\varrho}$ and of the unit propagation vector $\mathbf{u}$

$$\rho=\mathbf{u}\cdot\mathbf{\varrho}\cdot\mathbf{u}. \tag{7}$$

In the crystallographic coordinate system, Eq. (7) becomes, for KDP and ADP,

$$\rho=\rho_{11}(u_1^2-u_2^2).$$

Because of the small value of ρ_{11} and of the sample orientation ($u_1\simeq u_2$), the optical activity does not affect the measurement of d_{36} for these two materials. Two different orientations were used for the quartz samples corresponding to the following values of the optical rotation:

$$(010)\rho\simeq\rho_{11},\quad (011)\rho\simeq\tfrac{1}{2}(\rho_{11}+\rho_{33}).$$

Due to the small values of the thickness, and in addition for the (011) samples to the fact that ρ_{11} and ρ_{33} have opposite signs,[23] optical activity has no measurable effect on our Maker-fringe experiment: The calcu-

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lated maximum correction on $I_M(0)$ is 2%. This has been checked by the following two experimental observations:

(1) The same value of $I_M(0)$ was measured on three quartz samples (that is, for different orientations and thicknesses).

(2) No second-harmonic radiation polarized perpendicular to the crystallographic twofold axis could be detected.

B. Comparison with Other Published Measurements

As we mentioned above, there have been several determinations of the relative values for d_{11} (quartz), d_{36}(ADP), and d_{36}(KDP). We summarize the results in Table III and include the results of the present study.

In order to make a meaningful comparison between the quoted values, it is necessary to list the results according to the measurement technique as well as the wavelength of the fundamental beam. We consider first the results given by the Maker-fringe experiment, and secondly those of the phase-matching technique.

1. Maker-Fringes Method

Before starting detailed discussions, we want to emphasize the importance in this kind of measurement of the determination of the coherence length l_c. As shown by Eq. (5), an error Δl_c in l_c induces an identical fractional error in d: $\Delta l_c/l_c=\Delta d/d$.

Most frequently the coherence length is deduced from the spacing between minima or maxima of the first few fringes near normal incidence. In addition to the fact that a precise value of l_c requires an analysis over a large range of θ, the use of approximate expressions for Ψ may introduce substantial errors. For instance, taking

$$\Psi\cong(\omega L/c)(n_\omega-n_{2\omega})\cos\theta' \tag{8}$$

leads to a systematic error which increases with θ.

A much better procedure for $\theta\leq 25°$ is to deduce the coherence length from[24]

$$l_c\cong\frac{L(\sin^2\theta_n-\sin^2\theta_{n+1})}{4n_\omega n_{2\omega}}, \tag{9}$$

where θ_n and θ_{n+1} are external angles for two consecutive minima. As an example, for a 2-mm-thick sample of ADP, Eq. (8) induces an error of 5% at $\theta=15°$ and an error of 15% at $\theta=25°$, while the accuracy of the coherence length deduced from Eq. (9) is greater than 1%.

As we can see in Table III, there has been only one previous measurement comparing quartz, ADP, and KDP using a Nd^{3+} solid-state laser.[8,9] Our measurements are in agreement with the reported value for the ratio $d_{11}(Q)/d_{36}(KDP)$, but they are in disagreement for the case of $d_{36}(ADP)/d_{36}(KDP)$. However, a detailed reanalysis of the experimental data has led to

[24] R. C. Miller pointed out this useful relation.

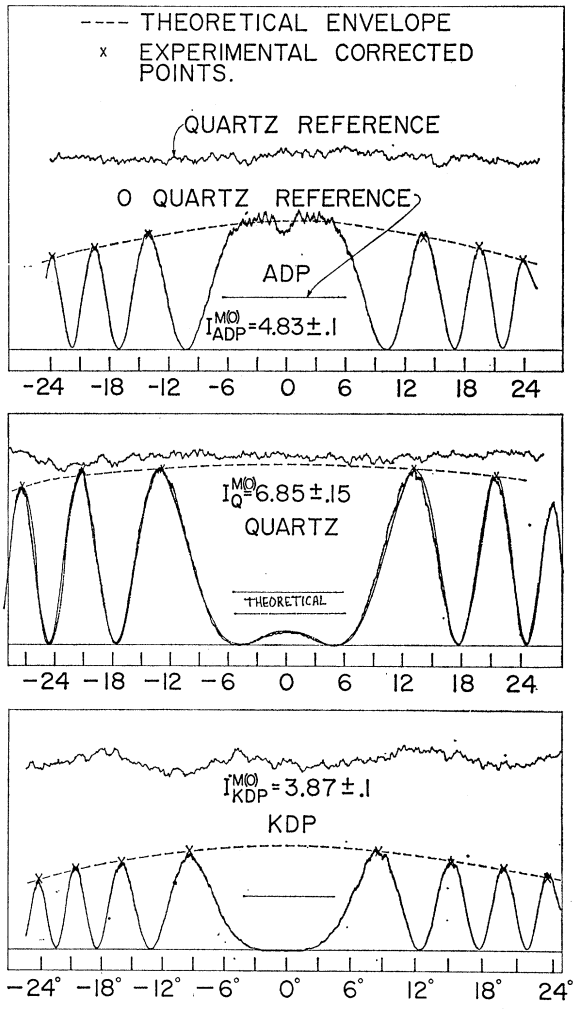

FIG. 1. Experimental Maker fringes are indicated for one sample of each material: quartz, ADP, and KDP. The crosses correspond to the maxima corrected according to the fluctuations of the second harmonic generated by the fixed quartz sample. The dashed lines are the theoretical envelopes. An example of the fit between theoretical and experimental fringes is given in the case of quartz.

the following corrected value[25]:

$$d_{36}(ADP)/d_{36}(KDP)=1.18\pm0.06,$$

and the discrepancy with the present results is thereby removed.

Because we did not perform any experiment using a ruby laser, the comparison with other results listed in Table III is less obvious. Nevertheless, all three materials are transparent in the near-uv range as well as in the visible and there should not be much variation, according to Miller's rule,[10] in the nonlinear susceptibilities.

As can be seen, our results are in fair agreement with those of van der Ziel.[11] It is interesting to note that the experimental values of l_c in this reference differ by only 1% from those deduced from Zernike's re-

[25] R. C. Miller (private communication).

fractive index data[19]: 9.7 μm for KDP and 9.2 μm for ADP.

Miller *et al.*[8] indicate a smaller value for d_{36}(ADP)/d_{36}(KDP), and a new analysis of the data does not lead to a good agreement[25]:

$$d_{36}(\text{ADP})/d_{36}(\text{KDP})=1.03\pm0.06.$$

Because of the uncertainty in the coherence lengths (experimental and calculated values reported in Ref. 12 are not in good agreement and differ from those deduced from Ref. 20), as well as uncertainties in the nonlinear susceptibilities, it is difficult to make any meaningful comparison with the data of Suvorov *et al.*[12]

2. *Phase-Matching Method*

From the experimental point of view, the Maker-fringe experiment and the phase-matching technique differ in many respects, the most important of which appear to be the following:

(1) The conversion efficiency is much higher in the phase-matching technique. As a consequence, experiments may be performed using cw lasers and absolute measurements are consequently less difficult.

(2) The structure of the fundamental beam (spatial intensity distribution, etc.) has to be known in phase-matching experiments for relative as well as for absolute measurements.

(3) The sample orientation with respect to the laser beam is much more critical in phase-matching experiments since the angular width of the second-harmonic peak is at least an order of magnitude smaller than the width of a Maker fringe for samples of identical thickness.

The accuracies of the two methods can therefore be quite different depending on the material studied and on the laser which is used.

Because of the small difference in wavelengths between the He-Ne and the Nd^{3+} YAG laser, there should be no significant variation of the nonlinear coefficients for ADP and KDP in the range 1.064–1.15 μ. Comparison of our results with those given by the phase-matching technique[13] indicates, however, an important difference (see Table III).

In order to determine the cause of this discrepancy, the author of the phase-matching experiment reanalyzed his data and took into account two previously neglected effects. Owing to the thickness of the ADP sample (2.5 cm), absorption is no longer negligible. In addition, the multiline He-Ne laser gave rise to an overlapping of the "harmonic" lines in the KDP crystal. As a result, the corrected value has been found to be [26]

$$d_{36}(\text{ADP})/d_{36}(\text{KDP})=1.03\pm0.16.$$

The present authors have also carefully studied the same phase-matching data.[27] The following three "fundamental" lines are involved in the "overlap" effect: 1.152, 1.161, and 1.177 μ. Their amplitudes can be estimated from the results on other samples (e.g., $LiNbO_3$, ADP) where the "harmonic" lines are resolved. It is then possible to perform a least-mean-squares analysis which indicates that the harmonic power at 0.572 μ generated by the KDP sample has been overestimated by 27±5%. Consequently, a corrected ratio has been obtained:

$$d_{36}(\text{ADP})/d_{36}(\text{KDP})=1.10\pm0.15.$$

Because of the difficulty in correcting for this "overlap" effect, a more definite answer to this important question will require a phase-matching experiment using a single-line laser.

CONCLUSION

The detailed comparison, including all the necessary refinements, of theory and experiment for the Maker fringe method has allowed us to obtain a precise determination of relative values for $d_{11}^{2\omega}$ (quartz), $d_{36}^{2\omega}$ (KDP), and $d_{36}^{2\omega}$(ADP). As a consequence of the corrections prompted by our work, the discrepancy among the previously published data has been largely removed. *Accurate* and *reliable standards* for optical nonlinear susceptibilities have thus been established.

The Maker-fringe method has been used for several years to measure the coefficients of almost all the known nonlinear optical materials. This work clearly demonstrates that a complete theory and careful analysis of the experimental data are necessary for accurate results. One must obtain a value of the coherence length which is in agreement with direct measurements of the refractive indices in order to deduce the correct value of $I_M(0)$. Otherwise, the accuracy is substantially poorer, as evidenced by comparing previous measurements for a given material.[1] Application of the complete Maker-fringe theory to earlier data has lead to corrected values in good agreement with those of the present study.

In addition, improved theoretical calculations of the nonlinear susceptibilities[28] provide a new stimulus for obtaining accurate and reliable experimental data.

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