III. RESULTS AND DISCUSSION

The absolute refractive index of strontium oxide for six wavelengths in the visible spectrum is given in Table I. The refractive index of air as given by Born and Wolf,[12] corrected to laboratory temperature and pressure, was used to refer the measured refractive index to vacuum. The probable error PE_n of n is the sum of the probable errors due to measurement of the refractive angle A and the angle of minimum deviation D. The differences in the refractive index of the two prisms are attributed to differences in crystal quality. Optical examination with a petrographic microscope revealed prism 2 to be of very high quality without strain. Prism 1 contained two distinct dislocations and appeared to be moderately strained. A spectroscopic analysis of the prisms was made to check the purity of the crystals. The following results were obtained: SiO_2, 0.03%; CaO, 0.5%; MgO, 0.1%; BaO, 0.5%; Al_2O_3, 0.1%; Fe_2O_3, 0.04%; MoO_3, 0.06%; MnO, 0.007%; NiO, 0.008%; CuO, 0.002%; Ag_2O, 0.0005%; and Cr_2O_3, 0.0001%. Analysis for carbon in the form of strontium carbide failed to reveal a trace (less than 1 part per million). The refractive index from 6600 to 4000 Å is shown in Fig. 3.

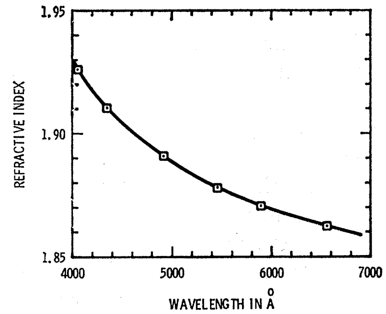

FIG. 3. The refractive index of single-crystal SrO in the visible spectrum.

[12] M. Born and E. Wolf, *Principles of Optics* (Pergamon Press Ltd., London, 1959), p. 95.

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cw Measurement of the Optical Nonlinearity of Ammonium Dihydrogen Phosphate

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We have measured the value of the element d_{36} of the nonlinear dielectric tensor of ammonium dihydrogen phosphate for the doubling of the 6328-Å line of the He-Ne laser. Two measurements were made. The first used a collimated single-transverse-mode laser beam (focal spot radius$\approx$0.2 cm) containing several longitudinal modes. The second used a weakly focused single-mode and single-frequency laser (focal spot radius $\approx$0.03 cm). In both experiments the crystal was slowly rotated through the index-matching angle. The value of d_{36} obtained by either method is $1.36\times10^{-9}(\pm12\%)$ in cgs esu. The experiments also verify the factor $(2n\text{-}1)/n$ for the enhancement of second-harmonic generation when n independent longitudinal modes are present.

INTRODUCTION

EARLY measurements[1,2] of optical nonlinearities were made by means of pulsed solid-state lasers. The limited transverse and longitudinal coherence of such lasers made the accurate interpretation of these experiments difficult. Ashkin *et al.*[3] first made a cw measurement in which the second harmonic[4] of the 1.15-μ line of a He-Ne laser was generated in a crystal of potassium dihydrogen phosphate (KDP). In their letter reporting this measurement, the authors mentioned the fact that the presence of several longitudinal modes with independent phases in the laser output enhances the harmonic generating efficiency of the laser beam by a factor $(2n-1)/n$ where n is the number of independently oscillating modes.[5] These authors further specify that the result of their measurement was not corrected for this effect because the number of longitudinal modes and the power distribution among them were not known. A cw measurement of the element d_{36} of the nonlinear dielectric tensor of ammonium dihydrogen phosphate (ADP) at 6328 Å was made by McMahon and Franklin.[6] Assuming the mode factor $(2n-1)/n$ to be approximately equal to two (for n large) they obtained the results: $d_{36}=2.0\pm0.5\times10^{-9}$ cgs esu. This is in fair agreement with the value $d_{36}=(3\pm1)\times10^{-9}$ cgs esu reported by Ashkin *et al.*, since this latter result was not corrected for laser multimoding.

[1] D. A. Kleinman, Phys. Rev. **128**, 1761 (1962).
[2] R. W. Terhune, P. D. Maker, and C. M. Savage, Appl. Phys. Letters **2**, 54 (1963).
[3] A. Ashkin, G. D. Boyd, and J. M. Dziedzic, Phys. Rev. Letters **11**, 14 (1963).
[4] Terminology introduced by P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23 (1963), for the frequency which equals twice the fundamental frequency.
[5] N. Bloembergen, in *Symposium on Optical Masers. Microwave Research Institute,* edited by Jerome Fox (Interscience Publishers, Inc., New York, 1963).
[6] D. H. McMahon and A. R. Franklin, Appl. Phys. Letters **6**, 14 (1965).

The present paper reports a similar cw measurement of the doubling of 6328-Å laser radiation in ADP, with considerable care given to the calibration of all factors in the experiment. Our result, corrected for the $(2n-1)/n$ factor, is $d_{36}=1.36$ cgs esu ($\pm 12\%$), or slightly lower than the previous measurements.[7]

In addition, we have performed these measurements using both a laser oscillating in multiple longitudinal modes and a single-frequency (single longitudinal mode) laser. Comparison of the harmonic generation obtained from these two lasers verifies the validity of the enhancement factor $(2n-1)/n$ for harmonic generation with multiple free-running longitudinal modes.

THEORY

The second-harmonic power generated by a laser beam with a single Gaussian transverse intensity distribution,[8] in a crystal of ADP oriented at the index-matching angle[9] is given by[3,10]

$$P^{2\omega}=2\frac{\omega^2 l^2}{\pi R_0^2}\left(\frac{\mu\mu_0}{\epsilon\epsilon_0}\right)^{3/2} d_{36}^2 P_0^2 \eta \sin^2\theta_m \qquad (1)$$

in mks units. The corresponding cgs expression is

$$P^{2\omega}=(128\pi^2\omega^2 l^2 d_{36}^2 P_0^2 \eta / R_0^2 n^3 c^3)\sin^2\theta_m\,, \qquad (2)$$

where n is the common index of refraction and c is the velocity of propagation of light in vacuum. Note that

$$d_{36}^{\text{mks}}=d_{36}^{\text{cgs}}\times 10^{-13}/27. \qquad (3)$$

The laser beam should enter the crystal as an ordinary ray. The harmonic will be an extraordinary ray. The symbols have the following meaning:

$P^{2\omega}$: harmonic power output,

ω: angular frequency of the laser,

R_0: spot radius of the Gaussian laser beam,[11]

l: length or "thickness" of the crystal in the direction of the laser beam,

$\mu_0/\epsilon_0=120\pi\Omega$: impedance of free space,

μ, ϵ: relative magnetic and electric permeabilities of ADP,

P_0: power in the laser beam,

θ_m: index-matching angle,[9]

d_{36}: element of the nonlinear dielectric tensor of ADP,

η: efficiency factor.[12]

Equation (1) assumes that the crystal is oriented for optimum second-harmonic generation in both the index-matching plane and the plane orthogonal to it. This means that the x and y axes of the crystal should make angles $\frac{1}{4}\pi$ with the plane determined by the axis of the laser beam and the optic axis of the crystal. Equation (1) also assumes that the length of the crystal is considerably smaller than the Rayleigh range[13] of the laser beam. Under this assumption the spot size of the beam is essentially constant throughout the crystal, as well if the crystal is located in the far field of the beam as when it is placed in the focus. The efficiency factor η accounts for imperfect matching of the indices due to the unavoidable spread of a laser beam of finite size. In the focus, where the spot radius is stationary and the beam parallel, the same factor η accounts for the loss of the harmonic conversion efficiency due to the effect of crystal anisotropy.[14,15]

EXPERIMENTS

Measurements were made on three ADP crystals of optical quality. These crystals were cut and polished in different thicknesses for near-orthogonal beam incidence at the index-matching orientation. Optical examination did not reveal any strains or multiple-domain structure. Further evidence that these crystals are indeed single crystals is derived from the fact that the width of the harmonic peak as a function of the angle of orientation near the index-matching orientation corresponds to the theoretical value. The presence of different domains would broaden this peak or would produce sidepeaks. The results obtained with each of the three crystals were in common agreement.

Two lasers were used: a Spectra-Physics Model 112 (near-hemispherical cavity 120 cm long and 8 mm in diameter) and a Model 119 (laser cavity 10 cm long and 1 mm in diameter).[16] Both lasers were operated in a single TEM_{00} Gaussian transverse mode. The Model 112 unit had an output of approximately 5 mW distributed over several longitudinal modes. The random, multi-frequency character of the intermode beats as observed using a silicon photodiode and an rf spectrum analyzer showed that the individual modes oscillated inde-

[7] In independent measurements carried out about six months later, using the same experimental apparatus, J. E. Bjorkholm has measured $d_{36}=1.38\times 10^{-9}$ esu $\pm 16\%$ under conditions of optimum focusing. All calibrations were independently repeated.

[8] G. D. Boyd and J. P. Gordon, Bell System Tech. J. **40**, 489 (1961).

[9] This is the angle between the optic axis of the crystal and the laser beam under index-matching conditions. See for instance: J. A. Giordmaine, Phys. Rev. Letters **8**, 19 (1962).

[10] R. H. Kingston and A. L. McWorther, Proc. IEEE **53**, 4 (1965).

[11] The spot radius is defined as the radius at which the electric field in the beam is $1/e$ times its value on the beam axis. The intensity at that radius is $(1/e)^2$ times the intensity on the beam axis.

[12] G. E. Francois and A. E. Siegman, Phys. Rev. **139**, A1 (1965); the efficiency factor η is given by curve I of Fig. 6 as a function of the normalized spread of the laser beam.

[13] The Rayleigh range is that part of the beam on each side of the focus where the intensity on the axis of the beam does not fall off by more than a factor of 2. [J. F. Ramsey, Space and Aeronautics R and D Handbook (1960–1961)]. It is given by: $l_R=\frac{1}{2}d^2k_0$ where d is the focal spot radius of the beam and k_0 is the propagation vector of the laser beam.

[14] G. D. Boyd, A. Ashkin, J. M. Dziedzic, and D. A. Kleinman, Phys. Rev. **137**, A1305 (1965).

[15] J. E. Bjorkholm (to be published).

[16] We are indebted to Spectra-Physics for the loan of this instrument.

pendently, without any mode-coupling effects. The power output of the shorter Model 119 was in the 100 μW range. Because of the short optical cavity and consequent large mode spacing, this laser was expected to oscillate in a single longitudinal mode only. This was evidenced by a clear observation of the Lamb dip.[17]

In order to measure the influence of the presence of several longitudinal modes, a relative measurement was made, comparing the results with the Model 112 and Model 119 lasers. We concluded that the longitudinal modes of the Model 112 were uncoupled and that the enhancement factor $(2n-1)/n$ was correct. Another relative measurement was also made by reducing the excitation on the Model 112. As the gain and the power of the laser tube went down, the harmonic efficiency decreased by a factor approximately two times higher than when a similar reduction of power was obtained by the use of neutral density filters attenuating the full multimode laser power.

The intensity distribution of the laser beam was very nearly Gaussian. The spot radius was measured by scanning a small detector (0.0025 cm diam) across the beam. The linear range of this detector extended at least two orders of magnitude beyond the light levels occurring in our measurements. The beam profile was found to be Gaussian to a good degree of approximation. In order to account for small local deviations from the ideal Gaussian intensity distribution, the same detector was scanned in a two-dimensional pattern across the beam. The resulting transverse profiles were digitalized and the equivalent Gaussian radius R_0 was obtained from the following expression:

$$R_0=[\Delta S(\sum I\Delta S)^2/\pi \sum (I\Delta S)^2]^{1/2}, \qquad (4)$$

where I is the local intensity in the beam and ΔS is the cross section to which I is attributed. This method gave essentially the same value for the radius as a direct examination of the quasi-Gaussian profiles.

The total power in the laser beam was measured by means of an Eppley thermopile No. 6087. This secondary standard was cross-checked with two similar units, Eppley No. 5389 and No. 5148 and was found to agree with both of the them within 1%.

The main problem of measuring the optical nonlinearity resides in detecting the weak second-harmonic output and calibrating the detector. The combination of a selected photomultiplier and a synchronous detector gave an equivalent noise input power at the harmonic frequency of 2.2×10^{-13} W in a bandwidth of 1 cps. Since the harmonic light levels occurring in our measurements ranged from 10^{-11} to 10^{-14}, we have obtained satisfactory signal-to-noise ratios by using bandwidths from 0.15 to 0.015 cps corresponding to integrating times of 1 to 10 sec. However, absolute calibration is still a major difficulty.

The photomultiplier (an RCA 6903) was of the type

[17] Professor A. Yariv (private communication).

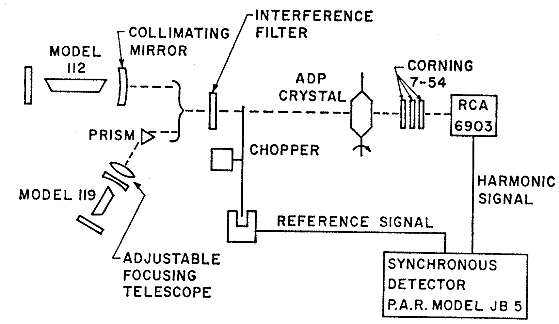

FIG. 1. Harmonic-generating experiment.

with flat, head-on, uv sensitive cathode, capable of intercepting the entire harmonic beam and insensitive to the polarization of light. An interference filter (and with the Model 119 laser a prism placed at the laser) served to eliminate the fluorescence of the He-Ne plasma. A series of 7–54 Corning glass filters was used to shield the detector from the fundamental beam (Fig. 1). The calibration was carried out as follows: At the time of each measurement the harmonic output obtained at the index-matching angle was compared to a small amount of fundamental power obtained from the laser beam by means of carefully calibrated attenuating filters. In this way the entire detection system, and in particular the photocathode and the associated electronmultiplier could be calibrated while operating under the exact same conditions pertaining to each experiment.

Before and after each series of measurements, the ratio of the quantum efficiencies of the photocathode at the fundamental and the harmonic frequencies was determined by a relative measurement. In this relative measurement, radiation at 3164 and 6328 Å was obtained from a high-pressure xenon arc lamp and a Leiss Model 9000 Prism Monochromator. The ratio of the energies emerging from the monochromator at the two wavelengths was determlned by means of a sensitive thermopile with fast response suitable for operation with the synchronous detector. The absolute calibration of this thermopile was not needed. Its neutrality was guaranteed by the gold coating which has a uniform low reflectivity ($<1\%$). Light levels suitable for this thermopile were still too high for the photomultiplier. Therefore, the output of the monochromator was attenuated by defocusing the arc lamp from the entrance slits and by moving the photomultiplier away from the monochromator to a distance such that it only could intercept a small part of the diverging output. We have verified that each of these operations was indeed neutral. The same central part of the photocathode which was active during the experiments was exposed in the calibration process.

Some additional attenuation was to be obtained from a reduction of the slit width of the monochromator. A reduction of the slit width reduces the power entering the monochromator and narrows the bandwidth of the

TABLE I. Typical experimental result with multiple-mode laser.

Laser: Model 112, operating in lowest order Gaussian transverse mode. Total power at the entrance face of the crystal 4.8 mW. Equivalent spot radius: 0.19 cm.
Power in the 6401-Å line: 0.3 mW.
Crystal thickness: 1.52 cm.
Harmonic power at the detector: 1.74×10^{-12} W.
Resulting value of d_{36}: 1.7×10^{-9} cgs esu (Typical result, not corrected for multimoding).
Average of repeated measurements: 1.85×10^{-9} cgs esu.

TABLE II. Experimental result with single-frequency laser.

Laser: Model 119. Monochromatic output power at the crystal: 105 μW. Focal spot radius: 0.033 cm.
Efficiency factor $\eta=0.70$.
Crystal thickness: 1.52 cm.
Harmonic power at the detector: 1.45×10^{-14} W.
Resulting value of d_{36}: 1.35×10^{-9} cgs esu.

apparatus. While the former effect is obviously neutral, the latter requires some consideration. In fact, this effect will be neutral only if the light source has no sharp intensity gradients in the region of interest. For this reason we have preferred the uniform and dense distribution of the lines of the xenon arc lamp over the higher power output at discrete lines of a mercury arc lamp. The low dispersion of the quartz prism which gives the monochromator a relatively large bandwidth (>50 Å in our experiments) justifies the use of the slit width as an attenuator for the xenon arc lamp.

In the course of our measurements it was observed that under certain operating conditions, the harmonic output as a function of the crystal orientation exhibited a secondary peak. Examination of the spectrum of the laser revealed that this peak resulted from the nonlinear mixing of a weak oscillation of the laser at 6401 Å with the main line at 6328 Å to give the sum frequency. The presence of a small amount of power at 6401 Å was taken into account as a reduction of the total measured laser output. Further corrections included reflections and absorptions at the crystal and the various filters.

Table I lists the different quantities as measured in a typical experiment using the Model 112 laser. Several experiments of this type were made over a period of several weeks and the resulting values of d_{36} ranged from 1.7 to 1.99×10^{-9} cgs, the average value being 1.85×10^{-9} cgs. These figures are not corrected for the effect of multimoding of the laser. Table II gives the results of a single measurement made with the Model 119 laser. This single-frequency laser was provided with an adjustable telescope and, in order to obtain a sufficiently strong harmonic output, we have weakly focused the beam in the crystal.

The value obtained for d_{36} in this measurement, which should be the "true" value, was $d_{36}=1.35\times10^{-9}$ cgs. This value must be compared with the multimode value of $d_{36}=1.85\times10^{-9}$ cgs, reduced by the multimode factor $[(2n-1)/n]^{1/2}$, where n is the number of modes oscillating. Unfortunately, we do not have any direct measurement of n for our laser, but experience with similar He-Ne lasers would indicate that n should lie somewhere between 5 and 15 modes oscillating. This gives a correction factor to d_{36} ranging between 1.34 for $n=5$ and 1.39 for $n=15$. The correction factor required to make our two experimental results agree is $(1.85/1.36)=1.36$, which would correspond to ~7 modes oscillating. Obviously, the correction factor is not very sensitive to the exact number of modes oscillating, and is close to the expected value.

The error limits for these measurements were estimated by estimating the percent error for each step in the calibration and measurement process and then adding these percentages (which presumably gives a somewhat pessimistic estimate of the cumulative error limits). The estimated error limits are compatible with the fluctuations in the experimental value from measurement to measurement.

CONCLUSIONS

We have measured the value of the element d_{36} of the nonlinear dielectric tensor of ADP for the doubling of the 6328-Å line of a He-Ne laser. Both a single-frequency and a multimode laser were used. Considering various possible experimental errors we propose the following result:

$$d_{36}=1.36\pm0.16\times10^{-9}\ \text{cgs}.$$
$$d_{36}=5.0\pm0.6\times10^{-24}\ \text{mks}.$$

This value is slightly lower than previous measurements of this quantity, although the stated error limits of this measurement and the earlier measurements do overlap.

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