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Second harmonic generation (SHG) and frequency mixing (FM) of infrared He-Ne laser beams in KDP crystal

Using an infrared He—Ne laser, which emitted four fundamental lines of wavelengths 1.153 μm, 1.161 μm, 1.198 μm, and 1.77 μm four second harmonic beams and six sum-frequency beams, generated in a KDP crystal (oriented at the phase-matching angle) were observed. The results obtained were in very good agreement with the theory of second harmonic generation and frequency mixing phenomena.

1. Introduction

In the 1960-ies, nonlinear optical effects were mainly observed with the use of strong light beams of ruby or neodymium: glass lasers. Second harmonics efficiencies from several to about 40 % were obtained in KDP, $LiNbO_3$, $BaNa_2Nb_5O_{15}$ and some other crystals, cut at an angle to satisfy the phase matching condition. Only very few papers were devoted to SHG with application of low power laser beams, such as He-Ne or other gas laser beams. ASHKIN, BOYD and DZIEDZIC [1] were the first to demonstrate, in 1963, the generation of the second harmonic, using 1.1526 μm He-Ne infrared lasers. This fundamental light beam, having a power of 1.48 mW, was focused in a KDP crystal. The second harmonic light beam emerging from this crystal had a power $8.1 \cdot 10^{-11}$ mW! Two years later MILLER, BOYD and SAVAGE [2], by applying much more efficient $LiNbO_3$ crystal and the so-called 90°-phase matching condition, succeeded in observing second harmonic light beams and frequency mixing processes. In the 90°-phase matching technique, the fundamental as well as the second harmonic travel perpendiculairy to the optical axis. The interaction was of the *oo-e* type. The spherical ordinary refractive indices surface (for fundamental beam) have to be tangent to the elipsoidal extraordinary refractive indices surface (for the second harmonic). To obtain this, the temperature of the $LiNbO_3$ crystal has to be raised to 193°C. The $n_1^0-n_2^e$ difference, which has a value of 0.011 at room temperature (for $\lambda = 1.153$ μm), decreases to zero at 193°C.

The He-Ne laser applied emitted four fundamental light beams (1.153 μm, 1.161 μm, 1,198 μm, and 1.177 μm). At phase matching condition, four second harmonic beams and six sum-frequencies were observed.

The theory of SHG and FM is due to FRANKEN and WARD [3], KLEINMAN [4], BOYD and KLEINMAN [5] and others. It gives a very good explanation of SHG and FM for unfocused as well as for focused light beams. SHG with the application of low power *cw* laser beams becomes very important since the new *cw* "pure" lasers have came to existence. The so-called pure laser contains neodymium pentaphosphate crystal (NdPP) with Nd^{3+} ion concentration of the order of 10^{22} cm^{-3}. In the experiment of CHINN [6], a $Ba_2NaNb_5O_{15}$ nonlinear crystal was placed within the optical resonator of an NdPP laser. When the laser fundamental beam ($\lambda = 1.05$ μm) was focused, internal second harmonic generation with an efficiency of 10 % was obtained. The power of fundamental light beam was but 1 mW! The NdPP laser was pumped with a 100 mW output dye laser. Thus, the total efficiency of the system was 0.1 %. It is surprising that such a weak laser beam is able to generate — via the crystal nonlinearities — a second harmonic light beam having a practically useful power level. Internal SHG in YAG:Nd^{3+} laser leads to spectacular power efficiencies approaching 100 % (see, e.g. GEUSIC and others [7]).

The aim of this work was to generate second harmonics and sum-frequency light beams with the use of He-Ne laser beams. Experimentally, very little attention was paid to these basic nonlinear phenomena.

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2. Measuring setup

A He-Ne infrared laser with a discharge tube of 120 cm in length and 3.2 mm in diameter was built. The discharge tube was filled with He^3 isotope to increase the output power. At a total pressure of about 6 Tr (He:Ne = 10:1), this laser emitted 20 mW infrared power in the four lines, namely 11 523 Å, 11 614 Å, 11 767 Å, and 11 985 Å. The discharge current was 17 mA. The light beam was filtered (to remove visible spontaneous radiation from the tube) and focused ($f = 5$ to 12 cm) within a KDP crystal, cut at an angle to match the phase velocities of the fundamental ordinary beam and the extraordinary second harmonic beam. The generated harmonics as well as the sum-freqencies entered the slit of a grating spectrograph — to measure their wavelengths (fig. 1). The infrared fundamental light beams were also observed at the exit diffused screen of this spectrograph using an image converter.
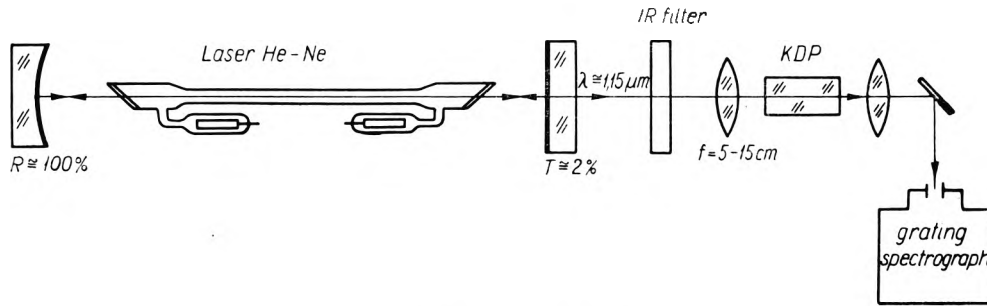
$oo \rightarrow e$ type interaction, two ordinary components of the fundamental beam (in the x and y-directions) generate one extraordinary component of the second harmonic. For the tetragonal symmetry, we have:

$$P_x^{2\omega} = 2d_{14}E_y^{\omega}E_z^{\omega},$$

$$P_y^{2\omega} = 2d_{14}E_z^{\omega}E_x^{\omega},$$

$$P_z^{2\omega} = 2d_{36}E_x^{\omega}E_y^{\omega},$$

where d_{14} and d_{36} are the only non-vanishing components of the nonlinear polarizability tensor. If the fundamental beam has components in the xy-plane, then only $P_z \neq 0$, whence we have:

$$P_z^{2\omega} = 2d_{36}E_x^{\omega}E_y^{\omega}.$$

For an arbitrary angle, φ, the $E_x^{\omega}E_y^{\omega}$ product is:

$$E_x^{\omega}E_y^{\omega} = (E^{\omega})^2 \sin\varphi\cos\varphi = \frac{1}{2}(E^{\omega})^2 \sin 2\varphi.$$

Fig. 1. Measuring setup

3. Preparation of the KDP crystals for SHG

Precise values of the refractive index of KDP as function of the wavelength were measured by ZERNIKE [8]. Using these values, the dispersion curve was plotted, enabling to calculate the refractive index for the second harmonic. Thus, the phase-matching angle was evaluated from the following well-known relation (see, e.g. KACZMAREK [9]):

$$\sin^2\Theta = \frac{(n_o^{\omega})^{-2} - (n_o^{2\omega})^{-2}}{(n_e^{2\omega})^{-2} - (n_o^{2\omega})^{-2}}, \qquad (1)$$

where ω and 2ω refer to fundamental and second harmonic, respectively. For $\lambda = 5761$ Å, $\Theta = 41°30'$, KDP crystals cut at this angle with respect to the optical axis were polished in the Central Optics Laboratory. Special quartz plates, attached by optical contact, protected their surfaces against humidity. These plates were antireflection coated to minimize the loss in power of the incident laser beam. The orientation of the crystal is illustrated in fig. 2. In
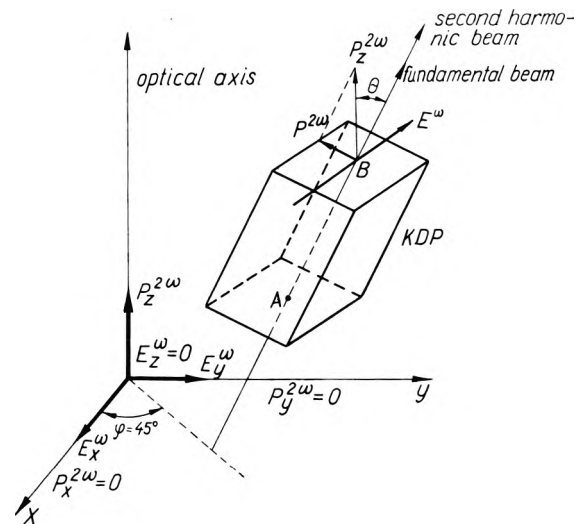

Fig. 2. Orientation of the KDP crystal with respect to the crystallographic axes and laser beam

For $\varphi = 45°$, this product becomes maximum:

$$E_x^{\omega}E_y^{\omega}(\varphi = 45°) = \frac{1}{2}(E^{\omega})^2.$$

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Thus,

$$P_z^{2\omega} = d_{36}(E^\omega)^2.$$

Optical polarization perpendicular to the direction propagation takes the following value:

$$P^{2\omega} = d_{36}(E^\omega)^2 \sin\Theta.$$

The intensity of SHG is proportional to:

$$d_{36}^2(I^\omega)^2 \sin^2\Theta.$$

The orientation of the crystal, as well as the electric field vector of the fundamental light beam, shown in fig. 2, ensure the maximum power efficiency in the process of SHG or FM, when the $oo\to e$ type of interaction is considered.

4. Frequency mixing

Frequency mixing can be described by the same relations as those used for SHG. Basically, there is no physical difference between SHG and FM phenomena. Frequency mixing was discovered by Bass et al. in 1962 [10]. Nonlinear polarization of the crystal can be written as follows (FRANKEN and WARD [3], BOYD and KLEINMAN [5], KIELICH [11]):

$$P_i^{(2)}(r, t) = \chi_{ijk} E_j^{(1)}(r, t) E_k^{(1)}(r, t). \qquad (2)$$

Let us consider two light beams of slightly different frequencies ω_1 and ω_2, incident on nonlinear crystal. Neglecting dispersion and vector notation, the formula (2) will be read as follows:

$$P^{(2)} = d(E_1^o)^2 \cos^2(k_1 r - \omega_1 t) +$$
$$+ d \cdot (E_2^o)^2 \cos^2(k_2 r - \omega_2 t) +$$
$$+ 2d E_1^o E_2^o \cos(k_1 r - \omega_1 t) \cos(k_2 r - \omega_2 t), \qquad (3)$$

where

$$E = E_1^o \cos(k_1 r - \omega_1 t) + E_2^o \cos(k_2 r - \omega_2 t),$$

and

$$\chi_{ijk} = d.$$

We have moreover:

$$P^{(2)} = \frac{1}{2} d(E_1^o)^2 + \frac{1}{2} d(E_2^o)^2 + \frac{1}{2} d(E_1^o)^2 \cos(2k_1 r -$$

$$- 2\omega_1 t) + \frac{1}{2} d(E_2^o)^2 \cos(2k_2 r - 2\omega_2 t) +$$

$$+ d E_1^o E_2^o \cos[(k_1 + k_2)r - (\omega_1 + \omega_2)t] +$$

$$+ d E_1^o E_2^o \cos[(k_1 - k_2)r - (\omega_1 - \omega_2)t]. \qquad (4)$$

We have obtained static polarization ("DC effect"), two second harmonic beams, sum-frequency and difference-frequency light beams. The respective intensities of these light beams are:

$$I(2\omega_1) = \frac{1}{4} A d^2 I_1^2,$$

$$I(2\omega_2) = \frac{1}{4} A d^2 I_2^2, \qquad (5)$$

$$I(\omega_1 + \omega_2) = \frac{1}{4} A d^2 I_1 I_2,$$

where A is a constant depending on the units used. Thus,

$$\frac{I(\omega_1 + \omega_2)}{\sqrt{I(2\omega_1)I(2\omega_2)}} = 4. \qquad (6)$$

It is remarkable that the intensity of the sum-frequency beam is four times larger that the intensity of the second harmonic beam. When the incident fundamental light contains several beams of different frequencies (e.g. infrared He-Ne laser used in our experiments), the sum-frequencies light beams are very easily detected among the slightly weaker second harmonic beams.

5. Experimental results

An infrared He-Ne laser emits several wavelengths in the vicinity of the 1.1523 μm main line. To analyse the spectrum, a grating spectrograph was used. Both spectra: that of the fundamental beams and that of the second harmonics and the sum-frequencies, were photographed. Fig. 3b shows an image converter picture of the infrared lines, and fig. 3a — the generated second harmonics and sum-frequencies. Taking the measured ν_1, ν_2, ν_3, and ν_4 values of the fundamental beams, second harmonic frequencies as well as the sum-frequencies were calculated. Agreement between the calculated values and those measured
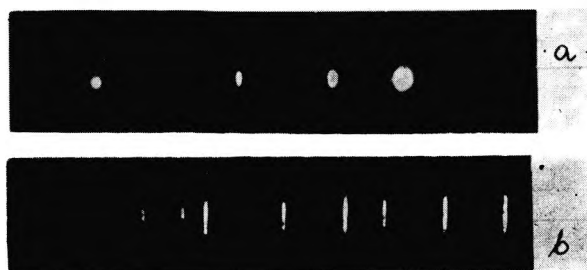

Fig. 3. Grating spectrograph spectra of the fundamental laser beams (a), and the harmonics and sum-frequencies (b). Picture (b) was obtained with the aid of an image converter

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with the aid of a grating spectrograph was very satisfactory. All the observed spectral lines, with the associated wavelengths, are plotted in fig. 4. The $2\nu_4$ second harmonics is not revealed in the picture, but it was distinctly observed visually. To calibrate the spectrum, mercury and sodium lamps as well as He—Ne red laser were used. When the fundamental laser beams were not focused, the harmonics and sum-frequencies were not resolved. A weak yellow-green light spot was observed in this case. Focusing of the laser beams ($f = 5$–12 cm) caused the almost circular spot to split into 10 well resolved colour lines (fig. 5). Elongation of these lines was perpendicular to the E-vector of the fundamental beams. As seen in fig. 2 and 5, this is the critical direction; thus, changes in the angle Θ cause a rapid mismatching of the phase velocities. On the other hand, changes of the angle φ are by no means critical, so that divergences of the focused beam do not disturb the phase matching condition. In this way, the linear shape of the harmonics (and sum-frequencies) expresses visually the influence of the phase matching technique on the intensity of the generated second harmonics and sum-frequencies.

Similar linear beam shapes of the ruby laser second harmonics was observed by AKHMANOW and KHOKHLOV [12]. Rotation of the crystal by angle of about 2° (with respect to the optimum phase matched angle, $\Theta = 41°30'$) caused the SHG and FM lines to disappear. The observed spectrum extended from yellow-green to orange-red. The maximum intensity for any desired line (colour) can be easily obtained by a small rotation of the crystal.

The measured difference $\Delta\Theta_{exp}$ in matching angles between the longest orange-red line (5934 Å) and the shortest yellow-green line (5761 Å) was $41' \pm 3'$. This difference was calculated using formula (1). The appropriate values of the refractive indices were evaluated from the dispersion curve. We obtained: $\Delta\Theta_{cal} = 41 \pm 5'$, the error being due to uncertainities in the refractive indices. Thus, agreement between these two results is satisfactory.

We have shown in this paper that a relatively low power laser beam of a He—Ne infrared laser can be successfully used to generate, in a nonlinear crystal (KDP or more efficient crystals like $LiNbO_3$, $Ba_2NaNb_5O_{15}$) several harmonics and sum-frequencies with very good visibility.
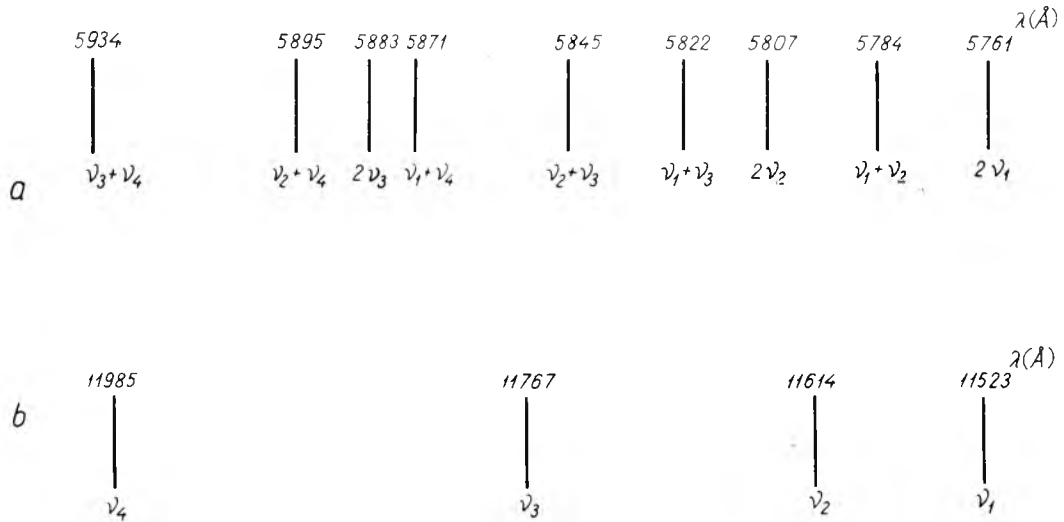

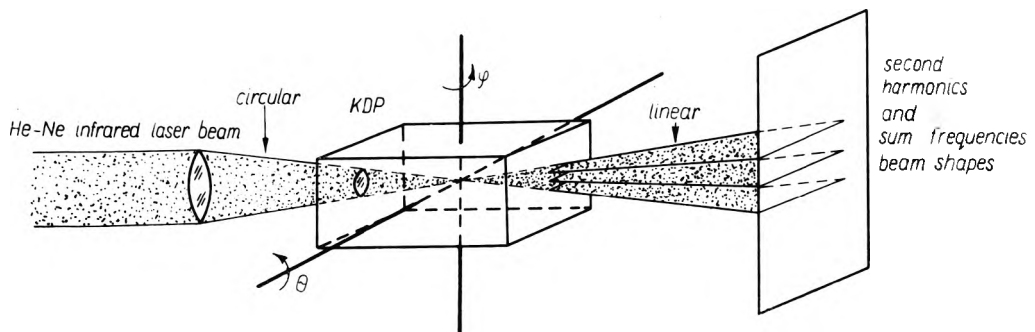Fig. 4. Illustration of the wavelengths, associated to the spectral lines obtained

Fig. 5. Orientation of the SHG and FM beam shapes with respect to the position of the KDP crystal

Acknowledgement

Генерирование второй гармоники и оптическое смешение инфракрасных пучков лазера He-Ne

Обсуждается наблюдение вторых гармоник и пучков света с суммовыми частотами (оптическое смешение) в кристалле KDP, расположенном под углом фазовой пригонки. Источником света был инфракрасный лазер He-Ne мощностью примерно в 30 мвт, испускающий четыре пучка света со следующими длинами волны: 1,153, 1,161, 1,198 и 1,177 мкм. Полученные результаты (четыре пучка вторых гармоник света и шесть суммовых пучков) находились в удовлетворительном соответствии с теоретическими расчетами.

References

[1] ASHKIN A., BOYD G.D., SAVAGE A., Phys. Rev. Letters **11**, 14 (1963).

[2] MILLER R.C., BOYD G.D., SAVAGE A., Appl. Phys. Letters **6**, 77 (1965).

[3] FRANKEN P.A., WARD J.F., Rev. Mod. Phys. **35**, 23 (1963).

[4] KLEINMAN D.A., Phys. Rev. **128**, 1761 (1962).

[5] BOYD G.D., KLEINMAN D.A., J. Appl. Phys. **39**, 3597 (1968).

[6] CHINN S.R., Appl. Phys. Letters **29**, 176 (1976).

[7] GEUSIC G.E., LEVINGSTEIN H.G., SINGH S., SMITH R.G., VAN-UITERT L.G., Appl. Phys. Letters **12**, 306 (1968).

[8] ZERNIKE F. Jr., J. Opt. Soc. Amer. **54**, 1215 (1964).

[9] KACZMAREK F., *Introduction to Laser Physics* (in Polish), PWN, Warszawa 1977

[10] BASS M., FRANKEN P.A., HILL A.E., PETERS C.W., WEINREICH G., Phys. Rev. Letters **8**, 18 (1962).

[11] KIELICH S., Opto-Electronics **2**, 125 (1970); *Molecular Nonlinear Optics* (in Polish), PWN, Warszawa-Poznań 1976.

[12] AKHMANOV S.A., KHOKHLOV R.V., *Problemy nelineynoy optiki*, ed. MGU Moskva 1964, p. 271.