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Phase matched doubly resonant third-harmonic generation in I₂-vapour

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Résumé. — A l'aide d'un laser pulsé utilisé en Rh6G pour produire la longueur d'onde fondamentale, nous avons enregistré le spectre d'excitation T.H.G. de la vapeur d'iode, en balayant la longueur d'onde du laser à colorant dans le domaine $5\,696 < \lambda_1 < 5\,820 \text{ \AA}$. La variation de la puissance de sortie V.U.V. avec la température de la vapeur saturante d'iode a été étudiée à $\lambda_{\text{V.U.V.}} = 1\,920,2 \text{ \AA}$. Nous avons déterminé la susceptibilité du troisième ordre $\chi^{(3)}$ pour cette longueur d'onde et la variation de la dispersion de la vapeur d'iode en fonction de la température.

Abstract. — Using the pulsed output from a Rh6G dye laser for the fundamental wavelength, we have recorded the T.H.G. spectrum of I₂-vapour, scanning the dye laser wavelengths between $5\,696 < \lambda_1 < 5\,820 \text{ \AA}$. The variation of the output V.U.V.-power with the saturation temperature of the iodine vapour has been studied at $\lambda_{\text{V.U.V.}} = 1\,920,2 \text{ \AA}$. The third-order susceptibility coefficient $\chi^{(3)}$ at this wavelength and the temperature dependence of the dispersion of the iodine vapour have been determined.

1. Introduction.

Frequency mixing and harmonic-generation have been carried out in the ultraviolet region using non-linear crystals. In the vacuum ultraviolet (V.U.V.), however, these crystals generally become opaque and can no longer be used. The techniques of Sum-Frequency Mixing (S.F.M.) and Harmonic Generation (H.G.) in atomic and molecular vapours [1] have attracted a great deal of attention as a means of extending coherent radiation sources to the V.U.V.-region of the spectrum. Recently, Tai *et al.* [2, 3] have used the technique of third-harmonic generation based on simultaneous resonances in one- and two-quantum transitions to find a level of symmetry $^1\text{O}_g^+$ of molecular iodine. They have shown that this technique is not as seriously limited by the photoionization process as the singly resonant method, and that it also has a high third-harmonic conversion efficiency. However, no details concerning the non-linear interaction process of the waves were given in their papers.

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In the present experiment we have recorded the emission spectra of the T.H.G. in I_2 -vapour and we have studied the temperature dependence of the V.U.V.-power i.e. on the phase-matching condition, determined the third-order susceptibility of I_2 in the region of $\lambda_{V.U.V.} = 1\,920\text{ \AA}$ and its dispersion $\Delta n = n(\lambda_{V.U.V.}) - n(3\lambda_{V.U.V.})$. We have also observed the $X \leftarrow F$ doubly resonant three photon excitation spectrum of I_2 in the region $1\,899\text{--}1\,940\text{ \AA}$.

2. Experiment.

A scheme of the experimental set-up is shown in figure 1. The light source is a Rh6G dye laser pumped by the S.H. of a pulsed YAG laser (Quantel). This Littman type dye laser constructed in our laboratory provided 10 ns long pulses with a bandwidth of $0.05\text{ \AA}$ at a repetition rate of 10 Hz. The laser power was approximately 0.4 mJ per pulse at $5\,760\text{ \AA}$. The laser beam was focused with a $f = 16\text{ cm}$ (or 10 cm) lens at the centre of a 10 cm long fused quartz cell containing I_2 -vapour. A Glan prism was put between the lens and the output mirror of the dye laser to ensure that the light was almost 100 % linearly polarized. Third-harmonic generation in the cell was separated from the laser beam by means of a V.U.V.-monochromator (Mc Pherson) and was detected by a solar blind photomultiplier (Model EMI G26 H315) which had a quantum efficiency of 13 % at $1\,950\text{ \AA}$. The output signal of the photomultiplier was integrated by a boxcar integrator (Model PAR 162/164) and recorded on a chart recorder. The gate width of the boxcar was 250 ns and the integration time constant $10\text{ }\mu\text{s}$. The dye laser wavelength was measured with an accuracy of $0.02\text{ \AA}$ using a Fizeau wave-meter (Laser Technics model 100) and an IBM micro-computer. A 100 MHz oscilloscope was used to control the time positions of the gate and the signal.

Fig. 1. — Schematic diagram of the apparatus, 1 : Rh6G dye laser. 2. Glan prism. 3 : Focusing lens. 4 : I_2 -cell. 5 : V.U.V.-monochromator. 6 : Solar Blind EMI-PM tube. 7 : Oscilloscope. 8 : Boxcar PAR 162/164. 9 : Recorder. 10 : Semi-reflecting mirror. 11 : Wave-meter. 12 : Computer. 13 : Photodiode.

In the first experiment the T.H. signal was recorded by scanning the dye laser wavelength in the region $5\,696\text{--}5\,820\text{ \AA}$. Figure 2 shows a part of this spectrum. To ensure that the generated signals were indeed due to T.H.G., either the I_2 -cell was cooled by liquid nitrogen or the fundamental beam was circularly polarized. In both cases no signal was detected.

The above-mentioned T.H.G. spectrum is due to simultaneous resonances in one- and two-quantum transitions [2]. Our spectrum is not part of the Cordes band system [5], or of the fluorescence spectrum reported by Kalatani *et al.* [6]. At room temperature ($20.5\text{ }^\circ\text{C}$), using focusing lens of 16 cm focal length at laser wavelength $5\,760.5\text{ \AA}$, one had a power of T.H.G.

Fig. 2. — Wavelength dependence of the generated T.H. power.

corresponding to 4.2×10^5 V.U.V.-photons generated per pulse, an energy conversion ratio of 1.0×10^{-9} .

The temperature dependence of T.H.G. power at the wavelength ($\lambda_{\text{V.U.V.}} = 1920.2 \text{ \AA}$) was investigated using two lenses of different focal lengths ($f_1 = 16 \text{ cm}$ or $f_2 = 10 \text{ cm}$). In the experiments the temperature of the I_2 -cell was varied continuously from 0°C to 20.5°C .

Figure 3 shows the variation of the T.H. power with temperature, i.e. with the number of iodine molecules per cm^3 [7]. From figure 3, it is obvious that the optimum T.H.G. power

Fig. 3. — Dependence of the generated V.U.V. power at $\lambda_{\text{V.U.V.}} = 1920.2 \text{ \AA}$ versus the temperature. (a) $f = 16 \text{ cm}$. (b) $f = 10 \text{ cm}$.

output will be reached above room temperature. Unfortunately, and investigation at temperatures higher than 21.°C was not possible in this experimental set-up.

3. Third-order susceptibility $\chi^{(3)}$ at the wavelength $\lambda_{\text{V.U.V.}} = 1\,920.2\text{ \AA}$ and dispersion Δn of the I_2 -vapour.

Theoretically, the magnitude of the third-order susceptibility $\chi^{(3)}$ of atomic gases can be determined by the perturbation theory. In the case of iodine vapour, results of ab-initio $\chi^{(3)}$ calculations are not yet available, and so we have used here the experimental value of $\chi^{(3)}$.

In the case of tight focusing of the fundamental wave, from [8] we have the formula

$$P_{\text{V.U.V.}}^{(3)} = \frac{12 (2\pi)^8 |\chi^{(3)}|^2 P_{01}^3 \sigma_3^2 \exp(-2\sigma_3) N^2}{c^2 \lambda_1^4 n_{\text{V.U.V.}}^3} \quad (1)$$

where $P_{\text{V.U.V.}}^{(3)}$, P_{01} are the powers of generated- and fundamental-waves respectively,

$$\sigma_3 = -\frac{b \Delta k}{2} \quad (2)$$

is the phase-matching parameter with

$$\Delta k = k_{\text{V.U.V.}} - 3k_1 = (6\pi/\lambda_1)(n_{\text{V.U.V.}} - n_1);$$

b is the confocal parameter of the laser beam; n_1 , $n_{\text{V.U.V.}}$ are the refractive indices at the wavelengths λ_1 and $\lambda_{\text{V.U.V.}} = \lambda_1/3$ respectively; N is the number of molecules per cm^3 ; c is the velocity of light in vacuo.

The confocal parameter is determined by an expression given by Kleinman [9]

$$b = 8\lambda_1 n_1^2 \left(\frac{f}{a} \right)^2 \quad (3)$$

where a is the diameter of the laser beam at entrance face of the focusing lens.

For constant values of temperature and wavelength, the ratio of the generated V.U.V. power using lenses of focal length f_1 and f_2 can be found from expressions (1) and (3), taking $n_1 \simeq 1$. This ratio is given by

$$\frac{V_{f_1}}{V_{f_2}} = \left(\frac{f_1}{f_2} \right)^4 \exp \left(\frac{48\pi(f_1^2 - f_2^2)}{a^2} \Delta n \right) \quad (4)$$

where V_{f_1} and V_{f_2} are the magnitudes of the corresponding averaged output voltage at the boxcar integrator. From equations (2), (3), (4) we have

$$\sigma_{3(f_1)} = -\frac{f_1^2}{2(f_1^2 - f_2^2)} \left(\text{Ln} \frac{V_{f_1}}{V_{f_2}} + 4 \text{Ln} \frac{f_2}{f_1} \right). \quad (5)$$

From (5), it is obvious that a and b are absent in the determination of the values of σ_3 .

At room temperature and at $\lambda_{\text{V.U.V.}} = 1\,920.2\text{ \AA}$, we measured

$$V_{(f_1 = 16\text{ cm})} = 7.8\text{ V}, \quad V_{(f_2 = 10\text{ cm})} = 2.7\text{ V}.$$

From equation (5) we obtained $\sigma_{3(f_1)} = 0.67$. Given that the peak power of our dye laser $P_{01} = 40$ kW produced a V.U.V.-peak power of $P_{V.U.V.} = 4.5 \times 10^{-5}$ W (4.2×10^5 V.U.V. photons/pulse) at room temperature ($N = 6 \times 10^{15}$ molecules/cm³), we obtained from (1)

$$\chi^{(3)} = 7.4 \times 10^{-34} \text{ e.s.u.}$$

This result shows that I_2 -vapour is a suitable non-linear medium for third-order sum-frequency mixing, like a metal vapour.

Using $a = 0.6$ cm, the curves (a), (b) in figure 3 and the expression (4) yield the value of the dispersion Δn . Figure 4 shows the dependence of $|\Delta n|$ on the temperature of I_2 -vapour. At room temperature, using $n = 1.001920$ [10], from the above results we obtained the refractive index of I_2 -vapour at wavelength $\lambda_{V.U.V.} = 1920.2$ Å and room temperature

$$n(1920.2 \text{ Å}) = 1.00192 \pm 1 \times 10^{-5}.$$

Fig. 4. — Dependence of the dispersion $|\Delta n|$ of the I_2 -vapour on the temperature (at wavelengths $\lambda_{V.U.V.} = 1920.2$ Å and $\lambda_1 = 5760.9$ Å).

4. Conclusion.

Upconversion of visible dye laser radiation by third-harmonic technique in iodine molecular vapour was investigated. We have determined the value of the third-order susceptibility $\chi^{(3)}$ of I_2 -vapour in the case of simultaneous resonance in one- and two-quantum transitions. We conclude that I_2 -vapour is capable of acting as a non-linear medium for third-order sum-frequency mixing.

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