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HAL Id: jpa-00210853
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Submitted on 1 Jan 1988

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Tunable V.U.V. radiation generated by non-resonant phase matched odd harmonic generation in xenon gas

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(Reçu le 20 mai 1986, révisé le 6 mai 1988, accepté le 15 juin 1988)

Résumé. — Du rayonnement V.U.V. accordable a été obtenu par génération du 5e harmonique dans le xénon et le domaine 1 135-1 170 Å en utilisant des faisceaux laser à colorant amplifiés. Dans le cas de la génération du (2m + 1)ème harmonique, sous la condition de focalisation forte, la valeur optimale du paramètre de phase adapté est égale à 2m - 1. Nous en avons déduit que la valeur de χ(5) du xénon est de l'ordre de 10^{-47} e.s.u.

Abstract. — Tunable V.U.V. radiation has been generated by non-resonant fifth-harmonic generation in xenon in the domain 1 135-1 170 Å using amplified dye laser beams. In the case of the (2m + 1)th harmonic, in tight focusing condition, the optimal value of the phase-matching parameter is equal to 2m – 1. We have deduced that the value of χ(5) of the xenon gas is of the order of 10^{-47} e.s.u.

1. Introduction.

In recent years the generation of tunable coherent radiation in the vacuum ultraviolet (V.U.V.) has been achieved in several gaseous systems by sum-frequency mixing and harmonic generation [1, 2]. These systems require careful attention with regard to phase-matching between the driving polarization and generated radiation. Bjorklund [3] has investigated the generation of ultraviolet radiation by the interaction of four waves in a non-linear isotropic medium. His calculations on the phase mismatch (defined as the difference between the wave vector of the generated radiation and the driving polarization) have been used for the interpretation of various experimental results [4, 5]. Several authors have investigated the cases of harmonic generation of higher order [6-9], however their results are not complete. In [6], Tomov and Richardson have given a theoretical analysis of fifth-harmonic generation (F.H.G.). In this work, only one value of the optimal focusing parameter was given. Reintjes has calculated the optimal value of the phase mismatch for special F.H.G. cases in which either the fundamental beam is focused into an infinite medium [7] or this beam is strongly focused [8] and in both cases, the experimental values of the powers of F.H.G. — wave are not determined concretely. In [9], theoretical and experimental results are not compared.

Fifth- and third-harmonic generation of the Nd:YAG fourth-harmonic in helium have been compared by Reintjes and She [10]. In their experiment the aperture was controlled by a transparent window at the entrance of the spectrometer so that the non-linear medium appeared to be semi-infinite, extending to one side of the beamwaist. Third harmonic generation and fifth-order resonant difference-frequency mixing in xenon in region 1 400 Å have been compared by Vallée et al. [11]. The results of two last references demonstrated that the higher-order process can be favoured by choice of resonant phase-matching condition.

In this paper, we investigate the generation of tunable coherent V.U.V.-radiation in a non-linear gaseous medium by non resonant phase matched harmonic generation and determine the optimal values of the phase mismatch required for optimum
V.U.V.-output. In order to achieve the high-power densities required by non-linear processes without damaging the windows of the cell containing the non-linear medium it is necessary to focus the fundamental beams tightly enough into the center of the gas cell, thus in this paper we discuss this case in detail.

In our experiments, fifth-harmonic generation (F.H.G.) of the fundamental output of a powerful narrow band dye laser have been carried out with xenon gas non-linear medium. We investigate the effect of the variation of the dispersion with the wavelength on the phase mismatch, consequently on the power of the generated V.U.V.-wave. Thereby, our theoretical results for the optimal value of the phase mismatch and the generated V.U.V.-wave for maximal power for F.H.G. in the wavelength region of negative dispersion between 1 135-1 170 Å have been verified experimentally. Moreover we have determined a value of the fifth-order non-linear susceptibility of xenon.

2. Theoretical analysis of odd harmonic generation.

In the isotropic non-linear medium, we have the following formula for the power of the generated \((2m + 1)^{th}\) harmonic wave [9]:

\[
P_{2m+1} = \frac{(2m + 1)^4 \pi^2 n_1^2 N^2}{C n_1 n_2 \omega_1^2} \times \left( \frac{k_1}{L} \right)^{2m+1} \frac{1}{b^2} \left| H_{2m+1}(\sigma_{2m+1}, \xi) \right|^2 \xi^{2m-2} \tag{1}
\]

where \(\chi^{(2m + 1)}\) is the \((2m + 1)^{th}\) order non-linear susceptibility of an atom ; \(\omega_1, n_1\) are the frequency and the refractive index of the medium at the frequency of the fundamental wave respectively ; \(k_1 = n_1 \omega_1 / C\) where \(C\) is the velocity of light in vacuum ; \(N\) is the number of the atoms in a cubic centimeter ;

\[
H_{2m+1}(\sigma_{2m+1}, \xi) = \int_{-\xi}^{\xi} \frac{e^{i\sigma_{2m+1} \tau}}{(1 + j\tau)^{2m+1}} d\tau \tag{2}
\]

where \(\xi = L/b\) is the focusing parameter, \(\tau = 2z/b\) where \(b = k_1 \omega_0^2\) is the confocal parameter and \(\omega_0\) the beamwaist of the fundamental wave,

\[
\sigma_{2m+1} = -\frac{\Delta k_{2m+1} b}{2} \tag{3}
\]

is the phase-matching parameter, with:

\[
\Delta k_{2m+1} = k_{2m+1} - (2m + 1) k_1 \tag{4}
\]

being the phase mismatch.

The function \(H_{2m+1}(\sigma_{2m+1}, \xi)\) is the focusing function which has its maximal value when the parameters \(\sigma_{2m+1}, \xi\) take the optimal values \(\sigma_{2m+1, \text{opt}}\) and \(\xi_{\text{opt}}\).

In the case of tight focusing, the function (2) has the form:

\[
H_{2m+1}(\sigma_{2m+1}, \xi \gg 1) = \frac{2 \pi}{(2m - 1)!} e^{-\sigma_{2m+1}} \tag{5}
\]

when

\[\sigma_{2m+1} > 0 \quad \text{i.e.} \quad b \Delta k_{2m+1} < 0 \tag{5'}\]

and

\[
H_{2m+1}(\sigma_{2m+1}, \xi \gg 1) = 0 \tag{6}
\]

when

\[\sigma_{2m+1} < 0 \quad \text{i.e.} \quad b \Delta k_{2m+1} > 0 \tag{6'}\]

Expression (5') shows that to satisfy the phase-matching conditions between the generated V.U.V. and the driving polarisation, the tuning range of the harmonic wavelength is restricted to the spectral regions with negative phase mismatch \(\Delta k (\Delta k < 0)\).

In the case of non resonance and tight focusing, for a given small region of the spectrum, because of the rapid change of the phase mismatch with the wavelength [5], the output power of the odd harmonic generated wave determined by (1) and (5) depends only on the phase-matching parameter \(\sigma_{2m+1}\). Thus in a concrete non-linear medium \((N = \text{const.})\) the power of the \((2m + 1)^{th}\) harmonic generated wave is maximal when the phase-matching parameter has the optimal value \(\sigma_{2m+1, \text{opt}}\). From (5) we see that the function

\[
H_{2m+1}(\sigma_{2m+1}, \xi \gg 1)
\]

has maximal values at the following optimum values of the phase-matching parameter

\[\sigma_{2m+1, \text{opt}} = 2m - 1 \, .\]

Figure 1 shows the dependence of

\[|H_{2m+1}(\sigma_{2m+1}, \xi \gg 1)|^2\]

on the phase-matching parameter in the case of tight focusing. Formula (7) yields:

\[
\sigma_{3, \text{opt}} (\xi \gg 1) = 1 \quad ; \quad \sigma_{5, \text{opt}} (\xi \gg 1) = 3
\]

and

\[
\sigma_{7, \text{opt}} (\xi \gg 1) = 5
\]

i.e. \(b \Delta k_3 = -2\) ; \(b \Delta k_5 = -6\) and \(b \Delta k_7 = -10\) .

(7)

This result is the same as that of Bjorklund [3] which requires that the non-linear medium should have a negative dispersion. The xenon gas satisfies the above condition and its dispersion changes rapidly with wavelength. We investigate the influence of this
Fig. 1. — Plot of $|\tilde{R}_{2m+1}((\sigma_{2m+1}, \xi = l)|^2$ as a function of $\sigma_{2m+1}$.

rapid change of the dispersion on the power of the fifth-harmonic generation in gas in experimental part.

Applying the results in [5, 12], with the wavelengths of the Rh610 laser beam and $b = 0.06$ cm, using the following formula:

$$\sigma_5 = \frac{b \Delta k_5}{2} = \frac{\pi b}{\lambda_{V.U.V.}} \left[ n(\lambda_{V.U.V.}) - n(\lambda_D) \right]$$

we have calculated the values of the refractive indices and of the phase-matching parameter $\sigma_5$ in the negative dispersion region (1172-1192 Å) of xenon gas under the pressure of 34.6 Torr (i.e. $N = 1.11 \times 10^{18}$ atoms/cm$^3$) (see Tab. I). In the other negative dispersion region of xenon gas (1135-1170 Å), using the condition as of above mentioned, the values of $n_{V.U.V.}$ and $\sigma_5$ are calculated and given in table II.

In the case of F.H.G., the equation (1) yields

$$P_{S,\xi = 1} = \frac{5(4)^7 \pi^{10} |\chi(5)|^2 P_{0D}^5 \sigma_5^2 e^{-2\sigma_5 N^2}}{9 \lambda_D^6 C^4 b^2 n_{V.U.V.} n_D}$$

Using the results in table II and $P_{0D} = 3.4$ MW, $\chi(5) = 2.4 \times 10^{-47}$ e.s.u. (see the experimental result in part 3 of this paper), $n(\lambda_D) - 1 = 0.703 \times 10^{-3}$ [12] and the parameter of (9) being determined previously, it was possible to obtain the curve representing the variation $P_5(\xi \neq 1)$ as a function of generated wavelengths $\lambda_{V.U.V.}$ (see Fig. 3). From this curve we see that the maximal value of the power of F.H.G.-wave is at $\lambda_{V.U.V.} = 1167$ Å, i.e. $\sigma_5 = 3(b \Delta k_5 = -6)$.

3. Experimental results.

In all the following experiments, we investigate the dependence of the output power of the fifth-har-
monic generated waves on the V.U.V.-wavelength (i.e. on the phase-matching parameter \(\sigma_5\)) in a small region using xenon gas as a non-linear medium. Because the spectral structure of this gas is relatively simple in comparison with that of molecular gases, it allows us to compare the experimental results with the theoretical results given above. Furthermore, with the powerful tunable narrow band TDL50 dye laser system in our laboratory the case of non-resonant phase matched frequency conversion could be investigated without saturation effects. Thus our experimental conditions are identical to those described in part 2 for the theoretical calculation.

3.1 EXPERIMENTAL SETUP. — The experimental setup is shown schematically in figure 2. The light source is a tunable dye laser system (model TDL50) pumped by S.H.G. of a YAG laser (Quantel) which provided 15 ns pulse with a bandwidth of approximately 0.01 Å and a repetition rate of 10 pulses per second. The linear polarized laser beam is focused with a \(f = 5\) cm lens at the center of a 10 cm long cell containing xenon gas at a pressure of 34.6 Torr (i.e. \(N = 1.11 \times 10^{18}\) atoms/cm\(^3\)). The diameter of the dye laser beam at the lens is \(a = 0.42\) cm giving confocal parameter \(b = (8 \lambda_D/n_D)(f/a)^2\) \([13]\) = 0.06 cm at the wavelength \(\lambda_D = 5\) 910 Å. The focusing lens is used at the entrance window of the xenon cell, the exit window is a 3 mm thick disc of MgF\(_2\) which has a transmission coefficient 0.042 at \(\lambda_{V.U.V.} = 1\) 135-1 192 Å. The generated signal is separated from the laser beams by means of a 80 cm Mc. Pherson monochromator and detected with a solar blind EMI G26H315 PM tube which has a quantum efficiency of 4 % and a gain of 10\(^6\) when it is operated at a voltage of 2 500 V. The output of the tube is recorded on an oscilloscope (Tektronic Model 456) with a bandwidth of 100 MHz.

3.2 GENERATION OF THE VACUUM ULTRAVIOLET RADIATION IN RANGES 1 173-1 192 Å and 1 135-1 170 Å. — In this section, we describe the experiment which perform the F.H.G. by focusing the fundamental laser \(\omega_D\) directly in xenon gas.

The Rh610 dye laser system operating in the fundamental TEM\(_{00}\) mode gives a constant output power of approximately 3.9 MW in a small spectral range (in the vicinity of 5 910 Å). The intensity of the generated V.U.V.-signal is deduced from the voltage of the PM tube taking into account the gain curve of the solar blind photomultiplier.

Taking into account the different reflexion losses for the V.U.V. signal (on the exit window of the xenon-cell, on the two mirrors, on the grating and on the two slits of the monochromator) we obtain the value of the output power of the F.H.G.-wave \(P_{V.U.V.} = 2.2 \times 10^6\) photons/pulse, i.e. 11.6 mW at \(\lambda_{V.U.V.} = 1\) 182 Å, which corresponds to a conversion coefficient of \(\eta = 3 \times 10^{-9}\).

Using the experimental value of the V.U.V.-output power \(P_{V.U.V.}\) with the equation (9) and the values of \(\sigma_5, n_{V.U.V.}\) in table I and \(N = 1.11 \times 10^{18}\) atoms/cm\(^3\) we calculate a value of \(\chi^{(5)}(1\) 182 Å) \(= 1.3 \times 10^{-47}\) e.s.u.

The experiments were repeated with the dye laser operating near \(\lambda_D = 5\) 830 Å and its power of 3.4 MW. We obtain an output power of the F.H.G.-wave \(P_{V.U.V.} = 12.9\) mW at \(\lambda_{V.U.V.} = 1\) 167 Å, which corresponds to a conversion coefficient \(\eta = 3 \times 10^{-9}\). The experimental results obtained by tuning the dye laser are shown in figure 3. The maximal output power of the F.H.G.-wave is at \(b \Delta k_5 = -6\) (\(\sigma_5(1\) 167 Å) \(= 3\), see Tab. II).

Repeating the calculation method given above, with the values of \(\sigma_5\) and \(n_{V.U.V.}\) in table II, the fifth-order non-linear susceptibility \(\chi^{(5)}\) is found to be \(\chi^{(5)} = 2.4 \times 10^{-47}\) e.s.u. at \(\lambda_{V.U.V.} = 1\) 167 Å. With this value the curve showing the power of F.H.G.-wave as a function of the V.U.V. generated wavelengths as expressed by (9) is shown in figure 3. Agreement between the corrected experimental values for the fifth-harmonic and the theoretical predictions is within a small factor which we feel to be reasonable. The uncertainty in the absolute experimental values is due mainly to the difficulty in determining the transmission of the monochromator.

4. Conclusion.

In this work we have obtained the theoretical expression for the output power of the odd harmonic generated wave in the case of tight focusing of the fundamental wave and we have determined the optimal conditions for the optimum value of the generated wave power. The arrangement in our laboratory has enabled us to investigate experimentally the dependence of the generated V.U.V.-wave output power on the wavelength. The agreement between experimental and theoretical results in the case of tight focusing in the center of the gas cell (this prevents the destruction of the gas cell windows) was satisfactory. Moreover the value of the fifth-order non-linear susceptibility near \(\lambda_{V.U.V.} = 1\) 167 Å has been determined.

Acknowledgments.

The authors would like to thank Professor Broyer for stimulating discussions and the loan of the experimental equipment.
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