MULTIPHOTON EXCITATION OF THE XENON NEAR THE $6s' |1/2|_1$ RESONANT STATE AND THIRD-HARMONIC GENERATION

Y. SALAMERO, A. BIROT, H. BRUNET, J. GALY, P. MILLET,
J.L. TEYSSIER and J.N. FOULQUIER

*Décharges dans les Gaz, Centre de Physique Atomique,
Université Paul Sabatier, UA-277, 118, Route de Narbonne,
F-31062 Toulouse Cedex, France*

The results obtained in a multiphotonic excitation of xenon near the $6s' |1/2|_1$ state by a tunable pulsed dye laser are presented. At low pressure the 1P_1 resonant state of xenon is created by 3-photon resonant absorption. When the pressure increases, the excitation spectra widen and shift towards shorter wavelengths. We explain this phenomenon by an excitation of the Xe_2 molecules in the ground state O^+g towards molecular excited state $1_u(^1P_1)$ correlated to 1P_1 state. A spectral analysis showed the presence in the luminescence of the first and second continuum of xenon. We demonstrated that the third-harmonic generation of the incident laser beam intervenes in the excitation process. This radiation was detected and its pressure dependence studied. The phase matching parameter C_{Xe} and the non-linear susceptibility were measured.

Experimental Set-up :

It has been described in previous paper (1,2). The excitation is achieved by a tunable laser pulsed beam (50 Hz). The dye used in the present experiment is the B.B.Q. centered at 388 nm. The energy of the laser pulses can be adjusted between 10 and 80 μJ . The bandwidth of the beam is about 0.8 cm^{-1} at 388 nm. This beam is focused in the gas by a 40 mm focal length lens. The luminescence is observed by a extremely solar blind P.M. set to detect single photon in a lateral direction to the incident laser beam. The third harmonic generation is detected in the frontal observation.

Lateral observation-Excitation and Emission Spectra :

At low pressure ($p_{Xe} < 1 \text{ torr}$) excitation spectra are very narrow and centered on the wavelength ($\lambda_{lr} = 388.7 \text{ nm}$) corresponding to the 3-photon resonant excited 1P_1 state (figure 1). When the pressure increases the excitation spectra widen and shift towards shorter wavelengths. Some failures appear in the excitation spectra (filtered at 129 ; 145 and 168 nm) at several reproducible incident beam wave

Figure 1 - Excitation spectrum of Xenon luminescence at 128 nm.

Figure 2 - Excitation spectrum of Xenon luminescence at 168 nm.

lengths (figure 2). This is interpreted by a molecular excitation of the free or bound dimers in the ground state towards the excimers $1_u(^1P_1)$ and/or $O_u^+(^1P_1)$. As the third-harmonic generation (T.H.G.) is present, these levels can be excited either by 1-VUV photon or 3-laser photon. Our results are in agreement with Miller and Compton experiment (3) who simultaneously detected T.H.G. and induced ionisation by the incident laser beam. The failures in the excitation spectra can be attributed, as shown by Aron and Johnson (4) at a 4-photon excitation process which increases the ionisation phenomenon. The fourth photon absorption intervenes probably between excited molecular states. Emission spectra shown only the first and second continuum of Xenon respectively centered at 150 and 170 nm. Their pressure dependance is identical to that obtained by a selective excitation near the 6s resonant state $3P_1(^1)$ or by other excitation means (5).

The presence of the 145 nm emission for pressures so weak than 1 torr, while the initial excitation is nearly resonant with the $6s' |1/2|_1$ state, traduces that a very quick process leads to the creation of the $6s |1/2|_1$ state. The $6p |1/2|_1$ and $6s' |1/2|_1$ states are strongly coupled by two-body collisions. The radiative de-excitation of the first conduces to creation of the 6s states (6).

The temporal analysis of the 145 and 170 nm emissions shows the participation of the $6s |1/2|_2$ (3P_2) metastable state to the creation of the molecular state responsible of the second continuum emission.

Third-harmonic generation : (T.H.G.).

When a laser beam is focused in a gaz with a wavelength λ_1 , the third-harmonic generation ($\lambda_3 = \lambda_1/3$) in the range of the negative phase mismatch ($\Delta k < 0$) is possible. The power of this VUV radiation generated is, starting from the expression of Ward and New (7) of his electric field :

$$P(3\omega) = \frac{1}{2} \epsilon_0 C \int 2\pi r |E(3\omega)(x^2+y^2)^{1/2}|^2 dr$$

we can write :

$$P(3\omega) = \frac{8.204 \cdot 10^{-16}}{(3\lambda_{03})^4} \chi(3\omega)^2 N^2 \cdot P_1^3 \cdot F(b \Delta k, b/L, f/L)$$

P_1 and $P(3\omega)$ are the incident and generated powers in W/cm^2 , b the confocal parameter, f the position of the focal point in the medium, L the length of the medium, N is in at/cm^3 , $\chi(3\omega)$ in $erg^{-1} \cdot cm^6$ (u.e.s.).

In the present experiment Δk is negative in the blue side of the resonance line at 129.6 nm ($^1P_1 \rightarrow ^1S_0$ transition). We have found by a calculation that the non-linear process is possible between 126.6 and 129.6 nm by application of the Sellmeir formula. We verified in the frontal direction the presence of this VUV tunable radiation. The VUV intensity is, like the luminescence, linear with the cube

Figure 3 - T.H.G. intensity (129.10 nm) versus Xenon pressure.

Figure 4 - Theoretical and experimental variation of C_{Xe} with VUV Wavelength.

of the incident laser beam energy. His bandwidth is very narrow, less than 10^{-2} nm. The pressure dependance of the T.H.G. intensity, for a laser excitation wavelength, presents a maximum situated at the optimal pressure (figure 3). The experimental points are suited by the theoretical law given by the $P^{(3\omega)}$ expression :

$$P^{(3\omega)} \sim AB^2 P_{Xe}^4 \exp(B \cdot P_{Xe})$$

The full curve of this figure is in good agreement with experimental points. From the determination of the optimal pressure ($b \Delta k = -4$) we found the phase matching parameter $C_{Xe} = \Delta k / N$. The figure 4 represents theoretical variation and experimental results of C_{Xe} versus VUV wavelength radiation. The disagreement in the wavelength range under 128.8 nm is probably due to a reabsorption of the VUV radiation when the pressure increases and by the intervention of molecules in the ground state whose population augments linear with the square of the pressure. The experimental determination of $\chi^{(3\omega)}$ has been possible by application of $P^{(3\omega)}$ expression. In table 1, we report several values of $\chi^{(3\omega)}$ for different wavelengths. Absolute error is about 100 % but the variation with the wavelength is significant. The measurements of the third-order non-linear susceptibility of Xenon in this range are not frequent.

$\lambda_{\text{laser}}(\text{nm})$	$\lambda_{\text{VUV}} = \frac{\lambda_{\text{laser}}}{3}$	$P_{Xe}(\text{torr})$	$\chi^{(3\omega)}(\text{cm}^6 \cdot \text{erg}^{-1})$
388.5	129.5	15	$1.3 \cdot 10^{-35}$
388.35	129.45	28	$1.0 \cdot 10^{-35}$
388.2	129.4	57	$6.7 \cdot 10^{-36}$
387.9	129.3	95	$4.9 \cdot 10^{-36}$
387.3	129.1	165	$2.8 \cdot 10^{-36}$
387.0	129.0	200	$2.0 \cdot 10^{-36}$

Table 1 - Different values of third non-linear susceptibility $\chi^{(3\omega)}(-\omega, \omega, \omega)$ of Xenon versus T.H.G. wavelength.

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