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Investigations of Third-harmonic Generation in Xenon and Argon by Means of a TOF Mass Spectrometer

Badania generacji trzeciej harmonicznej w ksenonie i argonie przy pomocy spektrometru TOF

INTRODUCTION

The knowledge of characteristics of the VUV light intensity resulting from the third-harmonic generation process in the isotropic media is very important for many experiments utilizing this process [1-10]. The aim of this work was to show the possibility of using an intersection of an effusive molecular beam with a focused photon beam, as an open ion source of TOF mass spectrometer for studying the third-harmonic generation in isotropic media. In the previous work the author presented investigations of third-harmonic generation in Xenon [10]. In this study the nonlinear medium were Xenon and Argon. The incident beam had wavelength of 3546 Å, and the generated radiation had wavelength of 1182 Å. A measure for the intensity of the generated VUV was the intensity of the ion current generated at the spot where a non-homogeneous effusive molecular (2,2-dimethylbutane) beam was crossed by the VUV beam. The influence of parameters like the diameter of the UV (incident) beam and Xe and Ar pressure on the VUV generation process was examined. The TOF mass spectrometer was used to analyse and register the ions being generated.

EXPERIMENTAL

The experimental set-up is shown in Fig. 1. The incident UV (3546 Å, 3.5 eV — photon energy) pulsed beam was generated by Nd: YAG Laser. Diameter of this beam was defined by the Diaphragm $D = 3$ mm and 7 mm.

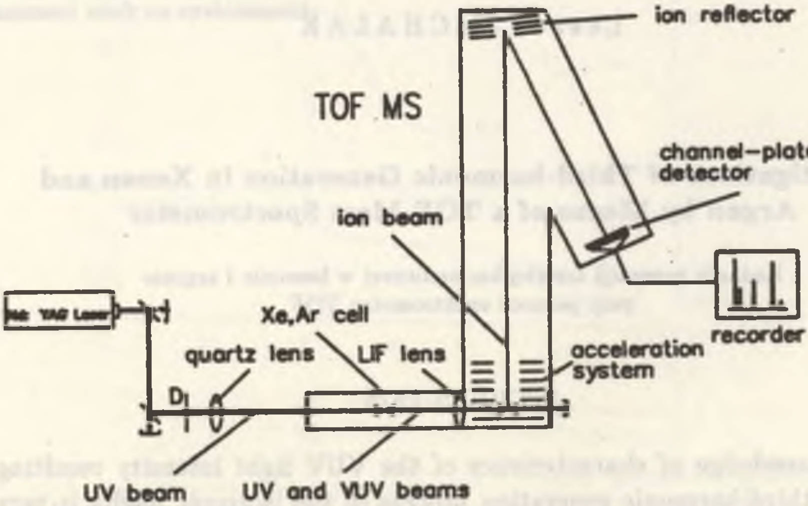

Fig. 1. Experimental set-up used for investigating third-harmonic generation in Xenon and Argon. The incident pulsed UV beam is generated by Nd: YAG Laser. A measure of the intensity of the generated VUV in the gas cell is the intensity of ion current from an ionization chamber TOF mass spectrometer. The PIMS (photoionization mass spectra) was recorded by using a channel plates detector and oscilloscope

The UV beam was focused in the center of the Xe and Ar cell (L =200 mm) by a quartz lens (f =200 mm). In the focus of the primary beam the vacuum ultraviolet (1182 Å, 10.49 eV — photon energy) was generated. The power density in the focus of the UV beam was varied by changing the size of the diaphragm D and had values 1.1×10^{11} W cm^{-2} and 1.4×10^{12} W cm^{2} for $D = 3$ and 7 mm, respectively. The pressure of Xe and Ar was monitored with a baratron. The LiF lens with the focal distance f_{UV} = 100 mm and f_{UVU} = 64 mm for UV and VUV respectively was placed after the gas cell and focused the VUV beam inside the ionization chamber of mass spectrometer and the same time formed the parallel UV beam.

On the place of the VUV focus a non-homogeneous effusion molecular beam was generated (perpendicularly to drafts in Figs. 1,2) by means of cylindrical capillary 70 mm long and $2R$ = 1 mm inner diameter (perpendicularly to drafts in Figs. 1, 2). A measure of intensity generated

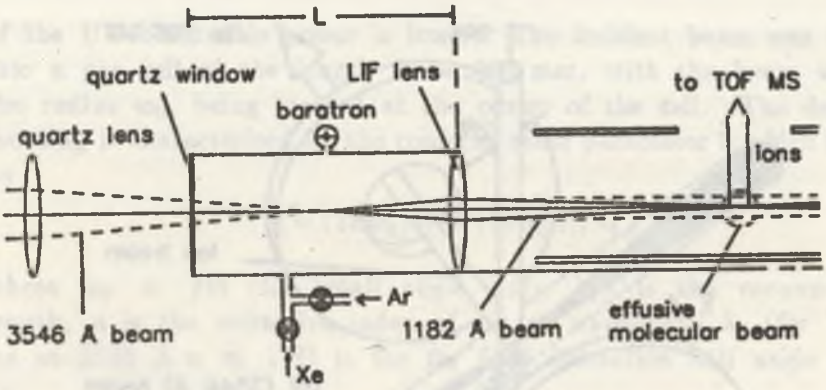

Fig. 2. The gas cell and optical system used for generation and detection of the VUV beam

VUV in the Xe cell was the number of 2,2-dimethylbutane (10.06 eV — ionization energy [11]) ions, which were generated in the intersection of the light and molecular beams and accelerated into the drift tube of the TOF mass spectrometer by an electric field. This system, where the effusive molecular beam is crossed with a photon beam just above the capillary outlet was the open ion source of this mass spectrometer. Using the non-homogeneous effusion molecular beam insured high density of molecules in the focus of VUV beam (higher numbers of produced photo-ions) [12–15]. Figure 3 shows the situation in the ion chamber of the mass spectrometer. The mass spectrum was recorded by using the channel plates detector and the LeCroy digital scope model 9400 with a sample rate of 100 MHz.

THEORY AND CALCULATIONS

The experimental investigations presented in this work were verified using the third-harmonic generation process theory in the case of very tight focusing near the center of the gas cell [16,17]. It comes from this theory that the power P_3 of the generated VUV beam is given by Equation (1)

$$P_3 = \frac{8.215 \times 10^{-2}}{(3\lambda_3)^4}[\chi(\lambda_3)]N^2 P_1^3 F(b\Delta k, b/L, f/L) \qquad (1)$$

where P_1 and P_3 are the incident and generated powers in watts, λ_3 is the wavelength of the generated beam in cm (for our case λ_3 = 1182 Å), $\chi(\lambda_3)$ is the third-order susceptibility at the third harmonic (for Xenon

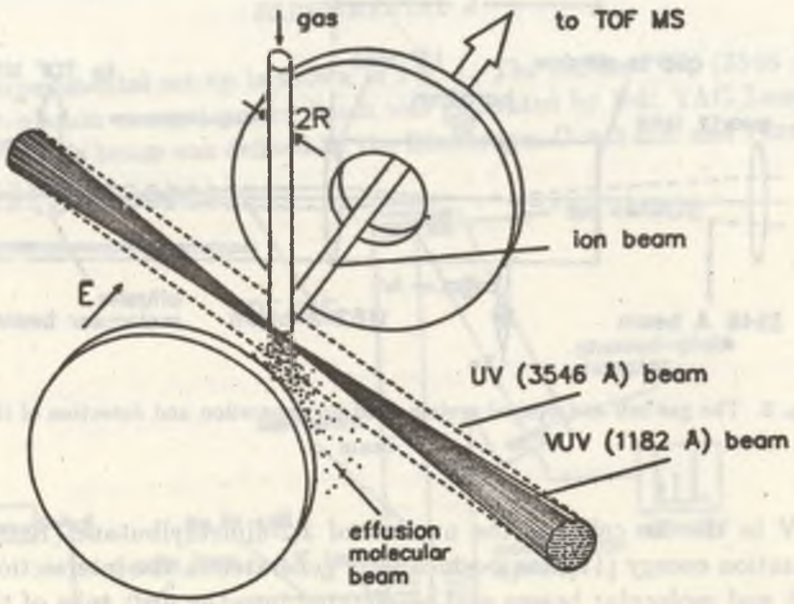

Fig. 3. The ionization chamber of the open ion source of TOF MS. The ions are generated in the intersection of the focused VUV and the effusive molecular beam

$\chi_{Xe}(1182\text{Å}) = 2.3 \times 10^{-35}$ esu [18]; for Argon χ_{Ar} is sufficiently smaller, that we can ignore its contribution to the total susceptibility of the gas mixtures [17]), N is the number density of the nonlinear medium in cm^{-3}, $F(b\Delta k, b/L, f/L)$ is a geometrical factor, b is a confocal beam parameter, L is the length of a cell which contained nonlinear medium, f is the focal length of the lens used for focusing of the incident (UV) beam, Δk is the wave vector mismatch defined by

$$\Delta k = k_3 - 3k_1 \tag{2}$$

where $k_3\vec{e}_4 = k_1\vec{e}_1 + k_1\vec{e}_2 + k_1\vec{e}_3$, $k_1\vec{e}_i$ and $k_3\vec{e}_4$ are the wave vector of the incident radiation with wavelength λ_1, and the generated radiation with wavelength λ_3 respectively. The incident beam is focused, so that in general the three unit vectors $\vec{e}_i$ come from three different directions and therefore k_3 is smaller than $3k_1$.

The knowledge of the incident and generated powers is not necessary for calculations of ratio P_3/P_1^3 but can be important for the interpretation of experimental results. In presented experiments, the pulsed (incident) UV laser beam had power 7×10^6 W for 7 mm (maximal diameter of UV beam used in our case) of the beam diameter. For smaller diameters of the UV beam this power is lower. The incident beam was focused into a gas cell of the length L = 200 mm, with the beam waist of the radius w_0, being located at the center of the cell. The degree of focusing is characterized by the confocal beam parameter b which is given by

$$b = (2\pi w_0^2)\lambda_1 = (2\pi w_0^2 n)/\lambda_{1,0} \tag{3}$$

where $w_0 = f\Theta$ (for small angle [19]), $\lambda_{1,0}$ is the vacuum wavelength, n is the refractive index of gas at wavelength λ_1 (for Xe and Ar at 3546 Å $n \approx 1$) Θ is the far field diffraction half angle defined as

$$\Theta = 0.5K(n_l)(D/f)^3 + 1.22\lambda/nD \tag{4}$$

where D is the diameter of incident beam, $K(n_l)$ is an explicit function of lens shape and n_l is the refractive index of the lens. For our case, where quartz (n_l = 1.55) plano positive lens were used, $K(n_l)$ is given by Equation (5) [19]

$$K(n_l) = \frac{1}{32}(n_l - 1)^{-2}(n_l^2 - 2n_l - 2/n_l). \tag{5}$$

In the tight focusing case ($b/L \approx 0$) the geometrical factor F has the following form [16]:

$$F(\Delta k, 0, 0.5) = \begin{cases} \pi^2(b\Delta k)^2 exp(b\Delta k/2), & \text{for } \Delta k < 0 \\ 0, & \text{for } \Delta k \geq 0. \end{cases} \tag{6}$$

In presented experiments b/L is 0.1 and 0.012 for D = 3 mm and D = 7 mm respectively but F differs not much for b/L = 0 [16]. Function F has a maximum for $b\Delta k = -2$. This can be obtained by changing the density N_{Xe} or N_{Ar} the Xe or Ar gas respectively, because

$$\Delta k = N_{Xe}C_{Xe}(\lambda) + N_{Ar}C_{Ar}(\lambda) \tag{7}$$

where $C_{Xe}(\lambda), C_{Ar}(\lambda)$ are the per atom dispersion of Xenon and Argon respectively (C_{Xe} (1182 Å) = -5.99×10^{-17}cm^2; C_{Ar} (1182 Å) = $+5.54 \times 10^{-18}$cm^2). From (1) using (3)–(7) we have calculated the ratio P_3/P_1^3 for diameters D = 3 and 7 mm of the incident (3546 Å) beam as a function of Ar pressure for several values of Xe pressure in the gas cell.

RESULTS

We can maximize the conversion by taking a pressure of Xe that is too high to reach a maximum conversion efficiency, and then adding a gas with normal (positive) dispersion until the right dispersion is obtained. The two factors N and F in (1) are now uncoupled and, in principle, one should be able to get a better conversion. A suitable gas to add is Ar.

Figure 4 presents conversion efficiency (P_3/P_1^3) of third harmonic generation as a function of Ar pressure for several constant values of Xe pressure, obtained for UV beam diameter $D = 7$ mm. An increase of generated power for higher values of Xe pressure and respectively higher Ar pressure is evident.

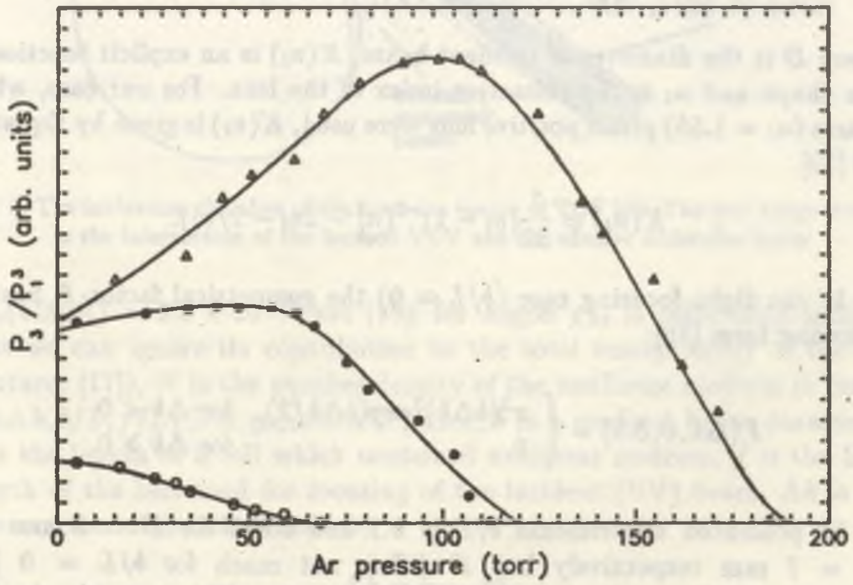

Fig. 4. Conversion efficiency $(P_3/P1^3)$ of third harmonic generation as a function of Ar pressure for Xe pressure $p_{Xe} = 5$ (○), 10 (•) and 15 torr (△). (—) theoretical calculated curve from (1). Diameter of UV (3546 Å) beam $D = 7$ mm

Figures 5 and 6 present normalized (at maximum values of the obtained power output of VUV) conversion efficiency (P_3/P_1^3) of third harmonic generation as a function of Ar pressure for several values of Xe pressure. Results presented in Fig. 5 and Fig. 6 were obtained for UV beam diameter $D = 7$ mm and $D = 3$ mm, respectively.

An increase in Xenon and Argon density does not infinitely increase the maximum conversion efficiency as (1) might suggest. At high densities the absorption of the generated radiation increase and even the inverse process of tripling can occur. Several authors have reported experiments in which limitations were observed in nonresonant harmonic conversion in rare gases [20]. Zych and Young [18] observed that the conversion efficiency for tightly focused beams in pure Xe reached a constant value of about 2×10^{-4}, becoming independent of the pump power for pump intensities above 5×10^{12} W cm^2. In our experiments the maximum value of the power density in the focus of the UV beam is 1.4×10^{12} W cm^{-2}.

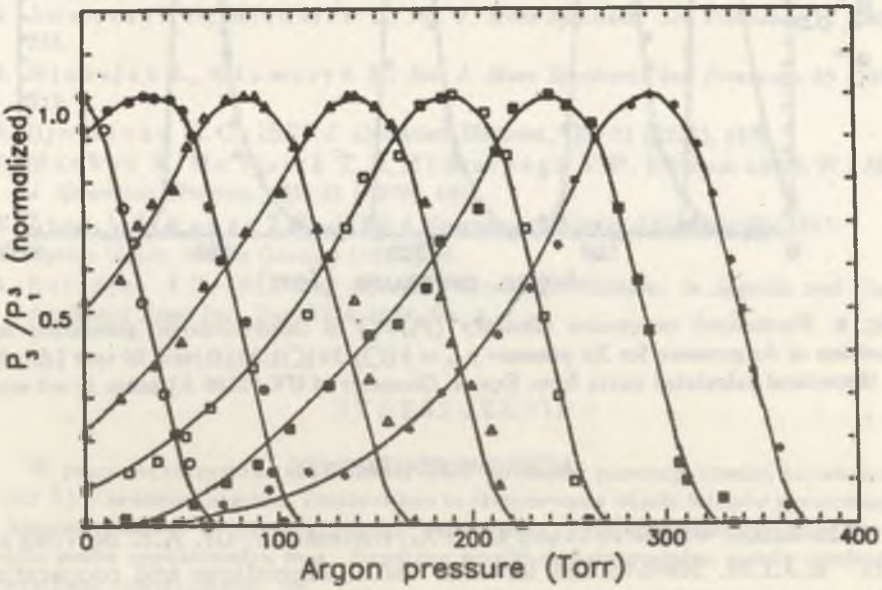

Fig. 5. Normalized conversion efficiency (P_3/P_1^3) of third harmonic generation as a function of Ar pressure for Xe pressure $p_{Xe} = 5$ (○), 10 (•), 15 (△), 20 (△), 25 (□), 30 (□) and 35 torr (*). (—) theoretical calculated curve from Eqn. 1. Diameter of UV (3546 Å) beam $D = 7$ mm

CONCLUSION

In this work, the system of crossing a non-homogeneous molecular beam with a focused VUV beam in an open ion source of a TOF mass spectrometer was used to investigate third-harmonic generation in Xenon and Argon. The influence the Xe and Ar pressure on the characteristcs of generation of the VUV beam was obtained. The good agreement between experimental results and the third-harmonic theory shows that the method used in this work appears to be very useful.

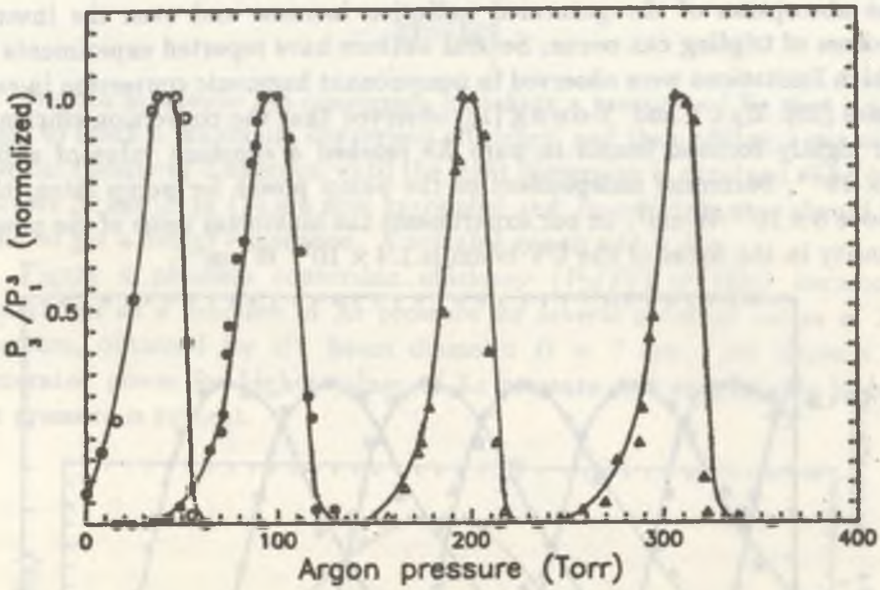

Fig. 6. Normalized conversion efficiency (P_3/P_1^3) of third-harmonic generation as a function of Ar pressure for Xe pressure $p_{xe} = 5$ (○), 10 (○), 20 (△)and 30 torr (△). (—) theoretical calculated curve from Eqn. 1. Diameter of UV (3546 Å) beam D =3 mm

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STRESZCZENIE

W pracy wykorzystano spektrometr TOF do badań generacji trzeciej harmonicznej (1182 Å) w xenonie i argonie. Zastosowano tu skrzyżowanie wiązki fotonów generowanych z komory gazowej z efuzyjną wiązką molekularną 2,2-dimetylobutanu jako otwarte źródło jonów spektrometru mas. Uzyskane wyniki eksperymentalne zostały porównane z wynikami teoretycznymi.

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STRESZCZENIE

W pracy wykonano [illegible] (THG) [illegible] ([illegible] Å) w [illegible] i argonie. [illegible]