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GENERATION OF COHERENT TUNABLE VUV RADIATION IN MERCURY

H. NKWAWO, C. MAÏNOS, M. AÏT-KACI, L. MUSEUR and M.C. CASTEX

Laboratoire de Physique des Lasers, Université Paris-Nord, Av. Jean-Baptiste Clément, F-93430 Villetaneuse, France

An experimental set-up for generating VUV light with use of non linear techniques in mercury is described. The effect of the buffer gas on a specific emission at 125 nm is discussed.

Third order nonlinear frequency mixing in a gaseous medium is a well established method for generating tunable coherent vacuum ultraviolet (VUV) radiation (1-5). To reach the region 122-127 nm where Kr atoms (123.6 nm) and Kr_n clusters absorb (6), the two-photon resonant four-wave mixing (TPFWM) in mercury vapor can provide VUV with a high conversion efficiency, up to 10% (5). Moreover it presents several experimental advantages due to the convenient dye lasers, doubling crystal and optics which can be used and because of the high vapor pressure of mercury provided at moderate temperature.

We describe here a quantitative analysis of VUV emissions produced by frequency mixing $\omega_{VUV} = 2\omega_1 + \omega_2$, when $2\omega_1$ is tuned to the two-photon transition $6s^1S_0 \rightarrow 7s^1S_0$ as illustrated by fig (1).

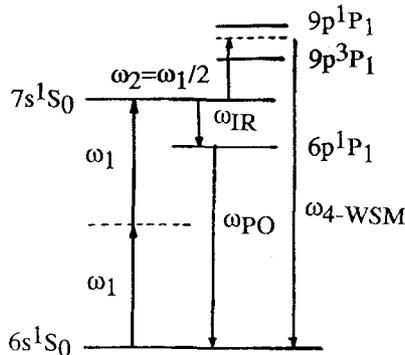


Fig. 1 : Energy diagram of Hg and typical mixing schemes .

To obtain tunable VUV light the experimental set-up used is illustrated by fig (2). A Nd Yag laser pumped two dye lasers simultaneously. The frequency of the dye laser 1 (Quantel TDL 50) is doubled by a KDP crystal and tuned to the two-photon resonance ($2\omega_1 = 63928 \text{ cm}^{-1}$). The frequency ω_2 of the dye laser 2 (home made) is tunable in the visible region. The two beams ω_1 and ω_2 , superposed using a dichroic mirror, transparent for the visible and Rmax for the UV at 313 nm, are focused into the mercury oven by an achromatic lens ($f = 28 \text{ cm}$).

The generated VUV is separated from laser lights by a VUV monochromator (90° stigmatic monochromator - Pouey prototype) and detected by a solar blind photomultiplier with CsI photocathode. The output signal of the photomultiplier is treated by a boxcar integrator (Par 162) and recorded by a microcomputer.

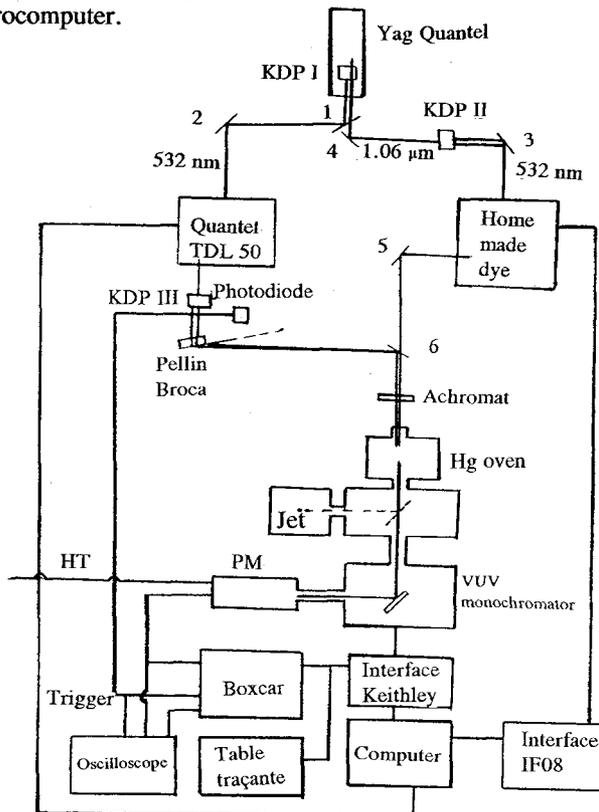


Fig. 2 : Experimental set-up for sum frequency mixing

With focused Gaussian beams, ω_1 and ω_2 , the intensity of the TPFWM VUV light is expressed by :

$$I_{VUV} = \text{const.} K_1^2 K_2 K_4 N^2 |\chi^{(3)}|^2 I_1^2 I_2 F(b\Delta k)$$

where N is the number density of Hg atoms, I_1 and I_2 are the intensities of dye lasers 1 and 2, $F(b\Delta k)$ is a phase matching factor which depends on the confocal parameter b and the wave vector difference $\Delta k = k_4 - 2k_1 - k_2$, $\chi^{(3)}$ is the nonlinear susceptibility which, not far from resonance, is given by:

$$\chi^{(3)} \propto \sum_{m,n} \frac{\langle 7s^1 S_0 | e_1 \mu | m \rangle \langle m | e_1 \mu | 6s^1 S_0 \rangle \langle n | e_2 \mu | 7s^1 S_0 \rangle \langle 6s^1 S_0 | e \mu | n \rangle}{(\omega_m - \omega_1) [\omega_n - (2\omega_1 + \omega_2)] (\omega_{7s} - 2\omega_1 - i\Gamma)}$$

where the quantities in angle brackets are electric dipole matrix elements connecting the states indicated, μ is the dipole moment operator and e_k is the unit vector defining the polarisation of wave k , Γ is the width of $7s$ level.

In order to optimize the VUV intensity which can be generated in our experiment, we have first examined, both experimentally and through simulation, the role of various parameters such as the two-photon detuning, the pressure broadening of the 7s level by the He buffer gas and the phase matching conditions for the Hg/He mixture.

For such an investigation, it was possible to use only the frequency doubled dye laser 1 (3 mJ at 313 nm and 0.2 cm^{-1} laser linewidth), the TPFWM process $2\omega_1 + \omega_1/2$ giving rise to an intense emission at 125.14 nm (7), as shown by Fig (3). Because of twofold enhancement by resonance with both $2\omega_1$ and ω_{VUV} ($9p^1P_1 : 125.05 \text{ nm}$), this emission was intense enough to be studied (i) at N_{Hg} densities as low as $3.10^{13} \text{ at/cm}^3$ and (ii) in spite of a positive Δk wave vector mismatch giving $F(b\Delta k) \sim 0$ for tight focusing conditions (8). Moreover, it was observed a strong decrease of VUV intensity as soon as the positive dispersive He gas, used as a buffer gas, was introduced in the Hg oven (fig.3), as well as a broadening of the 125 nm emission profile by He when $2\omega_1$ is tuned across the 7s level.

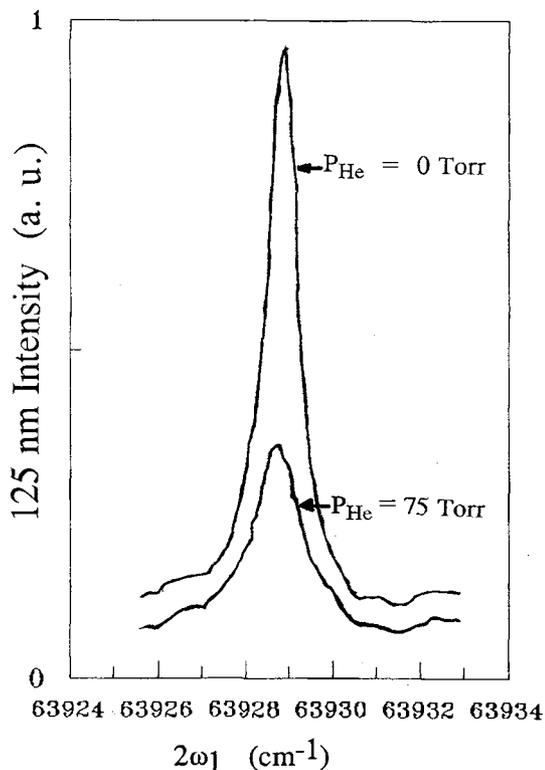


Fig. 3 : 125 nm emission when $2\omega_1$ is tuned across the 7s level for $N_{\text{Hg}} = 3.10^{13} \text{ at/cm}^3$.

To check the importance of this two effects, we have performed simulations of the observed VUV emission and calculated its intensity variation as a function of P_{He} . For the calculation of $\chi^{(3)}$, the experimental He collisional broadening ($5.10^{-3} \text{ cm}^{-1}/\text{torr}$) is included in the 7s Γ linewidth (natural Hg is used here). The $F(b\Delta k)$ function calculated depends on atomic densities: $b\Delta k = b(C_{\text{Hg}}N_{\text{Hg}} + C_{\text{He}}N_{\text{He}})$ where $C(\lambda)$ is the wave vector mismatch per atom.

As illustrated by fig (4), the results of our calculations are consistent with our experimental observations.

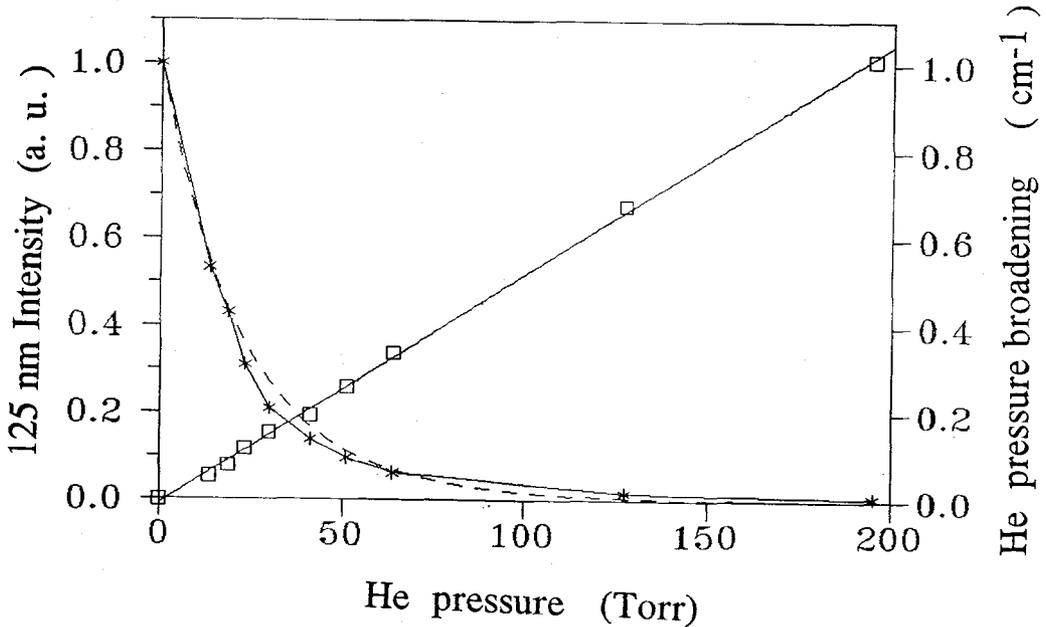


Fig. 4 : 125 nm intensity variation with P_{He} : x and □ experimental values
 ---- simulation

The intense 125 nm emission ($5 \cdot 10^{11}$ photons/pulse) obtained very easily (Hg cell is at room temperature) and with a high conversion efficiency (10^{-4}) can be a very useful emission for VUV spectroscopy.

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