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COHERENT VUV/XUV GENERATION USING TECHNIQUES OF NONLINEAR OPTICS : A SHORT REVIEW OF EXPERIMENTAL ACHIEVEMENTS

J. Lukasik

*Laboratoire d'Optique Quantique du C.N.R.S., Ecole Polytechnique,
91128 Palaiseau, France*

Résumé - Les dispositifs expérimentaux développés par les groupes de recherche à Palaiseau, Bielefeld, Chicago et Berkeley-Stanford sont brièvement présentés. Ils permettent, par les méthodes de l'optique nonlinéaire, la génération du rayonnement cohérent et accordable dans l'ultraviolet lointain et extrême. Le choix est représentatif du point de vue de particularité des dispositifs et des résultats obtenus.

Abstract - Experimental arrangements developed by research groups in Palaiseau, Bielefeld, Chicago and Berkeley-Stanford are briefly presented. They lead to the generation of coherent and tunable VUV/XUV radiation by nonlinear optical methods. The choice is representative from the point of view of the specificity of the setups and the results obtained.

Besides carbon monoxide (see the article by Vallée, Hellner and Lukasik), we were also interested in Palaiseau in xenon. As numerous other groups, we have produced the VUV radiation through the third order nonresonant sum and difference frequency mixing around 1150, 1400 and 1940 Å. The novelty of our experiments is, however, an efficient generation of the VUV light through a fifth order resonant difference frequency mixing : $\omega_1 + 3\omega_2 - \omega_2 \rightarrow \omega_{\text{VUV}}$ [1]. The fact that the difference frequency process is involved removes an important limitation connected with the dispersive region of the medium. It has been shown and proven [2] that under tight focusing conditions the sum scheme is restricted to negatively dispersive regions of the spectrum while the difference process may occur everywhere. The generated, through such a process, VUV radiation may thus be very widely tunable. The VUV powers obtained in these fifth-order difference frequency mixing experiments were of the order of 10 W in 8ns.

R. Hilbig and R. Wallenstein in Bielefeld showed in 1982 that xenon and krypton can be used as nonlinear media for the generation of intense, coherent VUV radiation, continuously tunable from 1100 to 2100 Å [3]. They described experiments involving sum- and difference - frequency mixing of the fundamental ω_L and the second harmonic ω_{UV} output of just one but powerful, narrowband dye laser in Xe and Kr. The sum-frequency $\omega_{\text{VUV}} = 2\omega_{\text{UV}} + \omega_L$ provides intense VUV radiation in the wavelength regions of negative dispersion between 1100 and 1300 Å. The required wavelengths of the fundamental laser operation (5500-6500 Å) are within the most efficient laser range of Nd:YAG harmonic pumped dye lasers. VUV radiation with the difference frequency $\omega_{\text{VUV}} = 2\omega_{\text{UV}} - \omega_L$ has been generated between 1850 and 2070 Å. Coherent VUV light of shorter wavelengths (1595 - 1866 Å) is obtained by mixing the UV dye laser radiation with the IR output ($\omega_{\text{IR}} = 1.06\mu$) of the Nd:YAG laser : $\omega_{\text{VUV}} = 2\omega_{\text{UV}} - \omega_{\text{IR}}$. In principle, the difference - frequency tuning range can be extended to wavelengths as short as 1226 Å and the method is thus covering continuously the entire range 1100 - 2100 Å. Powers quoted are in the range of few tens of W for a pulse of 5 ns.

A. Timmermann and R. Wallenstein just published another interesting result [4]. They report on the first known generation of single - frequency CW coherent VUV radiation.

The frequency of a stabilized dye ring laser (commercial system : Spectra-Physics model 380D) with the spectral width < 1 MHz is tuned to the two-photon resonance $3s^2\ ^1S_0-3s3d^1D_2$ ($\lambda_L=430.88\text{nm}$) of Mg. A laser power of only 0.2 W generated in a mixture of Mg and Kr third-harmonic ($\lambda_{\text{VUV}} = 143.6\text{ nm}$) VUV radiation of more than 1.2×10^5 ph/s ($1.8 \times 10^{-13}\text{W}$). This first experiment demonstrates that the third order frequency conversion of a CW laser light produces a useful amount of a narrow-band coherent VUV radiation. The authors expect a rapid efficiency improvement of the VUV source to about $10^7 - 10^9$ ph/s.

Some of the most spectacular experiments in the XUV generation and applications have been carried out over the past few years by the group of Professor Charles K. Rhodes at the University of Illinois at Chicago Circle [5-8]. This group has developed an ultrahigh spectral brightness ArF* source operating in the nanosecond (~ 7 ns) or picosecond (< 10 ps) regime. This originally designed system starts with a single frequency CW dye laser at about 5800 Å which is pulse amplified in a three-stage, XeF* pumped, amplifier. The resulting 10 ns, 20 mJ visible pulses are then focused into a Sr heat pipe making possible the third harmonic generation at 193 nm with pulses of 200 mW peak powers. These pulses are subsequently amplified in three discharge-pumped ArF* laser amplifiers, to produce 7 ns, 300 mJ pulses with the spectral width of the order of 0.01 cm^{-1} (< 260 MHz). The beam divergence is of only few μrad and the repetition rate 10 Hz. The tunability of such a source is achieved by setting the dye laser at the appropriate frequency within the ArF* gain profile (100 cm^{-1}). A similar system can be operated in the ps regime.

This source at 193 nm has been used to generate high spectral brightness, coherent tunable radiation in the vicinity of 64 nm by frequency tripling in various media (H_2 , D_2 , Ar, Kr and CO). A maximum harmonic output power of 30 W corresponded to a conversion efficiency of 10^{-6} . A frequency mixing scheme (two ArF* photons plus one visible dye laser photon) produced 200mW of tunable radiation in the vicinity of 79 nm. A similar KrF* system, in a frequency tripling scheme, can produce tunable, in the vicinity of 83 nm, XUV radiation.

The group in Chicago has used the above mentioned sources to the studies of collisional, radiative and spectroscopic properties of molecular hydrogen, deuterium and argon. Recently, stimulated VUV emissions on Lyman and Werner bands of H_2 following two-photon (193 nm) excitation to the E,F state have been observed [9-10]. Also, stimulated emission in the XUV originating from inner-shell transitions of atomic krypton was reported for the first time and described in ref. [11].

A.H. Kung of the SF Laser Center in Berkeley applied a novel method in nonlinear optical conversion scheme in which the nonlinear medium is produced in a pulsed supersonic jet [12]. This work was followed by a paper by Bokor and al. [13] of Bell Labs in which a similar technique, using a picosecond KrF* excimer laser tuned to 248.4 nm, led to the production of coherent radiation at 35.5 nm by seventh harmonic conversion. This is the shortest wavelength at which the generation of coherent radiation by nonlinear optical techniques has been reported in the open literature.

The most important technical problems that must be addressed in XUV generation experiments are the lack of suitable transparent window materials and self-absorption of the generated light by the nonlinear medium. These problems are usually overcome by employing a differential pumping system. The new jet method consists of confining the nonlinear medium to a small volume. The nonlinear interaction takes place at the intersection of the laser focus with an orthogonally directed, pulsed supersonic gas jet. Such an arrangement provides a localised region of high gas density in the vicinity of the nozzle orifice while substantially reducing the gas load on the pumping system. Another benefit is the reduced gas-consumption rate. The pulsed jet device resembles to an automotive fuel injector and is actually commercially available. The nozzles are of the order of 0.1 - 1 mm and the gas pulse duration, synchronized with a pulsed laser, is about 500 μs . In Bell Labs experiment helium was used as the nonlinear medium and its density at the interaction region is quoted

to be $2 \times 10^{17} \text{ cm}^{-3}$. With a 20 mJ, 15 ps KrF* laser the energy conversion efficiency to 35.5 nm is about 3×10^{-11} .

It is worth noting that the value of the pulsed supersonic jet technique was positively assessed in Berkeley-Stanford experiments [14] on quantum-state specific detection of molecular hydrogen using laser-induced fluorescence. The detection sensitivity of the system turned out to be about $2 \times 10^8 \text{ mol/cm}^3$ in a given ro-vibrational level of the ground electronic state of H_2 and the authors hope to lower it to 10^7 mol/cm^3 .

In conclusion, one may say that nonlinear optical techniques are today an established tool of production of coherent and tunable VUV/XUV radiation. Typically, 10^{11} – 10^{12} ph/s can be produced in such experiments. The bandwidth of this radiation is usually of the order of 0.1 cm^{-1} . With 10 ns pulses at the repetition rate of 10 Hz, a cone angle of about 10^{-4} rad corresponding to a beam with a Gaussian spot size of 1 mm, the average brightness is greater than $10^{24} \text{ ph/s/cm}^2/\text{\AA}/\text{sr}$ and the peak brightness in a single pulse is above $10^{31} \text{ ph/s/cm}^2/\text{\AA}/\text{sr}$. This high spectral brightness compares favorably with other VUV/XUV sources such as synchrotron radiation or arc lamps and is mainly due to a narrow linewidth of frequency converted laser sources and their excellent collimation.

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