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NARROWBAND TUNABLE VUV/XUV RADIATION GENERATED BY THIRD-ORDER FREQUENCY MIXING OF LASER RADIATION IN GASES

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Narrowband Tunable VUV/XUV Radiation Generated by Third-Order Frequency Mixing of Laser Radiation in Gases

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Third-order frequency mixing in gases is a well established method for the generation of optical radiation in the spectral region of the vacuum ultraviolet (VUV) at wavelengths λ_{VUV}=100-200 nm and of the extreme ultraviolet (XUV) at λ_{XUV}=58-100 nm. The pioneering work in this field has been published more than ten years ago by Ward and New[1], Harris and Miles[2], Miles and Harris[3] and Kung et al[4]. During the past decade the results of a large number of theoretical and experimental investigations demonstrated that frequency mixing of powerful laser light generates intense VUV and XUV radiation with fixed or tunable frequency and high spectral brightness[5-7].

Nonresonant frequency tripling and sum- and difference frequency mixing ($\omega_{VUV}=2\omega_1+\omega_2$) generated in the rare gases Xe, Kr and Ar broadly tunable VUV radiation in the wavelength range λ_{VUV}=110-200 nm[8-15]. Third harmonic generation in Ar and Ne produced XUV light in spectral regions between 72 and 105 nm[16,17].

In these experiments laser pulse powers of 1 to 5 MW provided conversion efficiencies of 10^{-5} to 10^{-6}.

The efficiency could be increased by several orders of magnitude by using resonantly enhanced frequency conversions[18]. By tuning the laser frequency to a two-photon resonance , for example, the induced polarization is resonantly enhanced. The two-photon resonant conversion, which is usually of the type $\omega_{VUV}=2\omega_1+\omega_2$, where ω_1 is tuned to a two-photon transition and ω_2 is a variable frequency, provides conversion efficiencies of $\geq 10^{-4}$ even at input powers of only a few kilowatts.

In the past the resonant frequency conversion has been investigated in metal vapors [19] (such as Sr, Mg, Cs, Ba, Hg and Zn) and in the rare gases Xe and Kr[20-22].

For experimental realization of the frequency mixing, rare gases are advantageous. Enclosed in a glas or metal cell (equipped with appropriate windows) these gases provide a nonlinear medium of homogenous, easily variable density. These gases are thus an appropriate medium for the construction of a reliable VUV light source useful for spectroscopy applications.

For spectroscopic applications parameters like the tuning range, the output power and the bandwidth are of special interest. In the following typical values of these parameters are summarized.

Tuning range

Phase-matching conditions between the generated VUV and the focused laser light restrict the tuning range of the sum frequency to spectral regions of negative mismatch ΔK, defined as the difference between the wavevectors of the generated radiation and the driving polarization[8,23].

The rare gases Ne, Ar, Kr and Xe provide the required negative dispersion at the high energy side of their resonance transitions

in extended regions of the wavelength range λ_{VUV}=66-147 nm (see Fig.1). Above the transmission cut off of LiF (λ_{VUV}=105 nm) Kr and Xe are negative dispersive in the wavelength regions 110-116 nm (Kr), 113-117 nm (Xe), 117.2-119 nm (Xe), 120-135 nm (Kr), 126-129 nm (Xe) and 140-147 nm (Xe). At these wavelengths nonresonant third harmonic generation and sum frequency mixing has been investigated in detail[3-7]. Frequency tripling of the third harmonic of the Nd-YAG laser - that generates intense VUV at 118.3 nm - and the generation of radiation at L_α=121.6 nm are well-known examples.

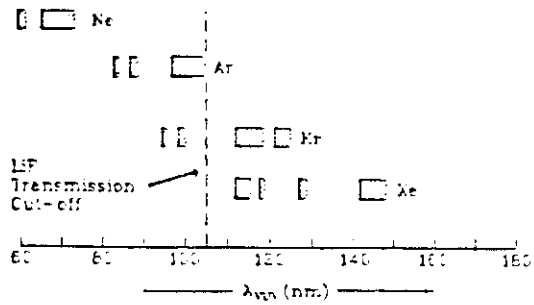

Fig. 1 Spectral regions with negative mismatch ΔK for frequency tripling and sum-frequency mixing

In contrast to the sum, the difference frequency can be generated in a medium with positive or negative mismatch[8-11]. Since this conversion is not restricted by the dispersion of the medium it should be suited for the generation of VUV in the entire range between 115 and 200 nm.

This has been demonstrated by mixing the fundamental frequencies of the dye laser (ω_1) or of the Nd-YAG laser (ω_{1F}) with the UV radiation with the frequency $\omega_{UV}=2\omega_1$ in the rare gases Xe, Kr and Ar[7,10,11]. Fig.2 displays the tuning ranges of the difference frequencies $\omega_{VUV}=2\omega_{UV}-\omega_{1F}$ (with $\omega_{UV}=2\omega_1$ and λ_1=420-700 nm) and $\omega_{VUV}=2\omega_{UV}-\omega_1$ (with $\omega_{UV}=2\omega_1$ and λ_1=420-630 nm)

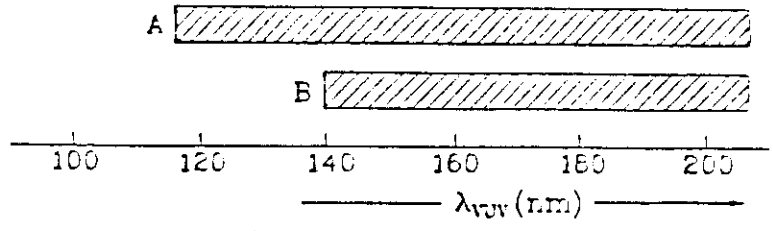

Fig. 2 Tuning range of the nonresonant difference frequency mixing in rare gases
A: $\omega_{VUV}=2\omega_{UV}-\omega_{1F}$ with $\omega_{UV}=2\omega_1$ and λ_1=420-700 nm
B: $\omega_{VUV}=2\omega_{UV}-\omega_1$ with $\omega_{UV}=2\omega_1$ and λ_1=420-630 nm
λ_1 is the wavelength of the dye laser.

It should be emphasized that this nonresonant frequency conversion requires only one dye laser pumped by the second or third harmonic of a Nd-YAG laser.

As the nonresonant difference frequency mixing the two-photon resonant difference frequency conversion produces widely tunable

VUV. In Kr, for example, the difference frequency $\omega_{VUV}=2\omega_R-\omega_T$ of UV radiation at $\lambda_R=212.5$ nm (resonant with the 4p-5p[1/2,0] two-photon transition) and of tunable dye laser light at $\lambda_T=210-900$ nm generates radiation at $\lambda_{VUV}=120-215$ nm (see Fig.3).

It should be emphasized that the resonant sum-frequency $\omega_{VUV}=2\omega_R+\omega_T$ generates continuously tunable radiation at $\lambda_{VUV}=72.2-96.7$ nm[20]. In this case the frequency ω_T is tuned at $\lambda_T=217-900$ nm.

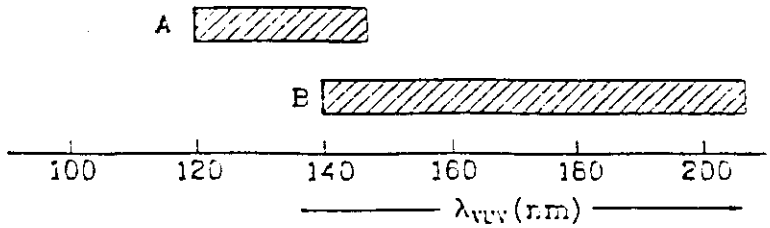

Fig. 3 Tuning range of the resonant frequency mixing in Kr: $\omega_{VUV}=2\omega_R-\omega_T$ with $\lambda_R=212.5$ nm. A: $\omega_T=\omega_1$, B: $\omega_T=2\omega_1$ ω_1 is the fundamental frequency of the dye laser ($\lambda_1=420-900$ nm).

Output power

In nonresonant frequency conversion laser pulse powers of 1-5 MW provide conversion efficiencies of 10^{-5} to 10^{-6}. The power of the generated VUV light pulses is typically in the range of 1-20 W ($0.3-6\times10^{16}$ photons/pulse). The efficiency is limited by dielectric gas breakdown in the focus of the laser light and by nonlinear intensity-dependent changes of the refractive index[23].

The resonant frequency mixing usually provides conversion efficiencies of 10^{-2} to 10^{-4} depending on the resonant enhancement of the nonlinearity and on the laser power. With input pulse powers of the resonant UV radiation of only 100-200 KW the VUV output power generated in Kr is in the range of 0.2-2 KW ($6\cdot10^{11}-6\cdot10^{12}$ photons/pulse)[20]. Higher input powers will further increase the VUV output by at least one order of magnitude.

Bandwidth

The spectral bandwidth of the VUV radiation is determined by the linewidth of the pulsed laser radiation.

The linewidth of commercial pulsed dye laser systems is typically 0.2-0.5 cm^{-1}. With additional line narrowing - provided by intracavity etalons - the spectral width is reduced to 0.02-0.1 cm^{-1}. For those conversions which use the radiation of the fundamental of the Nd-YAG laser injection-seeded systems may be used. The linewidth of these lasers is less than $3\cdot10^{-3}$ cm^{-1}. For narrowest bandwidths the radiation of cw dye lasers is amplified in pulsed dye laser amplifiers. The fourier-transform-limited linewidth of the amplified radiation is also less than $3\cdot10^{-3}$ cm^{-1}.

Depending on the used laser systems the bandwidth of the generated VUV radiation is on the order of 0.01-1.5 cm^{-1}.

Because of the small spectral width the spectral brightness of narrowband VUV generated by resonant frequency conversion, for example, is on the order of 10^{17} photons $sec^{-1}nm^{-1}$. This value surpasses the spectral brightness of synchrotron VUV light sources (which is typically 10^{14} photons $sec^{-1}nm^{-1}$) by orders of magnitude.

The discussed results demonstrate that nonlinear optical frequency conversion produce widely tunable VUV radiation. Because of the narrow spectral width and the high intensity the VUV light is a powerful tool for high resolution spectroscopy of atoms and molecules. This has been demonstrated, for example, by absorption spectroscopy, by excitation spectroscopy or by state selective resonant-excitation-ionization spectroscopy (see ref. 5-7 and references therein).

Today the number of spectroscopic applications of coherent laser-generated VUV light is still small. Because of the simple way of generation and the excellent spectral properties there is no doubt that in the future this radiation will be very useful for a large variety of spectroscopic applications.

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