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MULTIPLE RAMAN SCATTERING SCHEME FOR XUV GENERATION

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Résumé - On propose un système avec double scattering stimulé de Raman et un résonateur à multipass pour la radiation IR. Le système est fondé sur la conversion dans la région XUV utilisant des niveaux quartet métastable dans l'afterglow d'une discharge He-K. On peut avoir des impulsions XUV à 64nm avec énergie 1,2μJ en utilisant un laser à colorant à 404nm de 14MW, 6ns avec une efficience ~ 1.7×10^{-4}.

Abstract - A scheme with double stimulated Raman scattering and multipass resonator for intermediate radiation is proposed based on up-conversion into XUV region by quartet metastable level of potassium atoms excited in a He-K discharge afterglow. 64nm wavelength XUV output pulse with 12μJ energy may be obtained with 404nm dye-laser input power of 14MW and 6ns duration with overall efficiency ~ 1.7×10^{-4}.

I - INTRODUCTION

Stimulated anti-Stokes Raman scattering (SASRS) processes have been found of some interest for achieving laser action in the VUV and XUV spectral regions /1/. Up-conversion of existing powerfull lasers' radiation to a shorter wavelength by SASRS on metastable atomic states was proposed long ago /2/, but only recently interesting experimental results were reported /3,4/. The problem is to find a suitable medium with a metastable level which can be pumped easily to achieve population inversion between the initial and the terminal levels. A tunable pump laser must be used to obtain resonance enhancement of SASRS /5/. An attractive scheme was proposed by S. Harris /1,6/ in which a terminal level may be selected which is not the ground atomic state. In this aproach a high-lying quartet metastable state of an alkali metal atom is populated by direct electron impact excitation in a discharge and the pump laser radiation is tuned to an intermediate doublet autoionization quasi-metastable state from which the lasing transition takes place terminating on an usual excited level of the alkali atom. However it turned out to be difficult to obtain the required initial metastable density in a pulsed discharge /7/. This difficulty may be overcome by using collisional energy transfer from rare gas atom metastable states to the quartet state of an alkali metal /8,9/. One of the proposed reactions is the following

$$He^*(1s2s\ ^3S) + K(3p^6 4s\ ^2S) \longrightarrow K^{**}(3p^5 3d4s\ ^4P_{5/2}) + He(1s^2\ ^1S) \qquad (1)$$

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with energy defect 0.04eV. Recent spectral measurements of 60-80nm wavelength emission from He-K discharge showed effective excitation of the potassium quartet states /10/. A powerful source of tunable IR (λ_{IR}=1.209μm) radiation is needed to pump SASRS in resonance with potassium quartet ("storage" level)- intermediate transition: $3p^53d4s\ ^4P_{5/2}$ - $3p^5(4s4p\ ^3P)\ ^2P_{3/2}$, (Fig.1 and /9/). In this way λ_{XUV}=64nm radiation may be produced in resonance with $3p^5(4s4p\ ^3P)\ ^2P_{3/2}$ - $3p^64p\ ^2P_{3/2,1/2}$. The optical oscillator strength of the pumping transition is $f_{si}=1.2\cdot10^{-3}$, and of radiating transitions f_{ie}=0.05 for J=3/2 terminal sublevel and 0.02 for J=1/2 /11/. Application of the pump radiation in afterglow conditions, i.e. delayed by 50-150ns from an actual discharge pulse, results in a decrease of the terminal 4^2P level population due to its radiation and an increase of the He 2^3S metastable density due to recombination processes, thus achieving the population inversion needed for SASRS. In this paper we consider a new possibility to obtain λ_{IR} =1.209μm radiation for SASRS pumping by means of preliminary Stokes Raman scattering (SRS) in the same He-K discharge. In this way the preliminary step is the stimulated SRS from He 2^3P level which in a strong violet radiation field (p=404nm) undergoes a transition to He 3^3P level producing λ_{IR} =1.209μm in resonance with He 5^3D level (detuning $\approx$80cm^{-1}). The measured density of initial He 2^3P atoms in pulsed discharge of [He] : [K] =4:1 mixture is high enough $10^{12}cm^{-3}$ /12/.

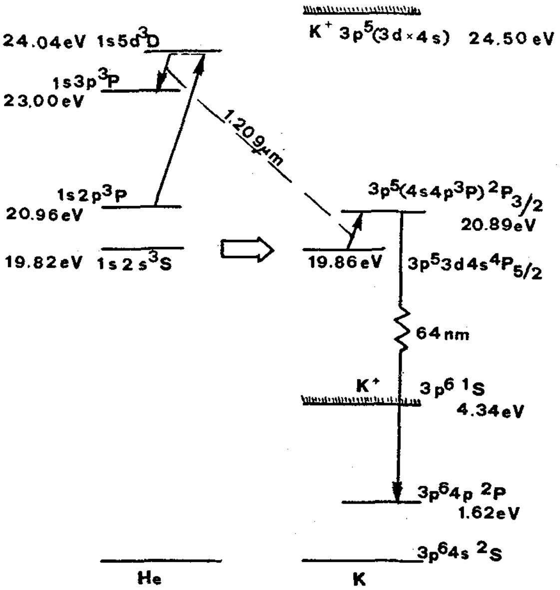

Fig.1- Energy level diagram for the proposed double Raman scattering scheme in the He-K system.

The second main point is to use high-quality IR mirrors for a multipass Raman cell /13,14/ so achieving a conversion efficiency from p=404nm to IR up to 20-30%. Calculations show that overall conversion efficiency of the double Raman process from the violet to XUV can reach several percents according to focusing geometries choosen.

II - INFLUENCE OF FOCUSING ON UP-CONVERSION OF THE IR RADIATION INTO THE XUV

The intensity of low-signal SRS radiation from the noise level is exponentially growing with distance x as exp $(g_{AS}x)$. The plane-wave gain coefficient for the resonant SRS in a three level system is given by /5/:

$$g_{AS}=\alpha_R N_S I_S \ (\mathrm{cm}^{-1}), \qquad (2)$$

where N_S is the storage $^4P_{5/2}$ state density, I_S ($\mathrm{W\,cm}^{-2}$) is the pump wave intensity (Stokes $\lambda_s=1.201\mu$m wave in ourscheme), and

$$\alpha_R = C\,\frac{f_{si}\,f_{il}\,\omega_{AS}}{\omega_s\,\omega_{il}\,(\Delta\omega_{AS})^2\,\Gamma_{AS}} \quad (W^{-1}cm^4) \qquad (3)$$

where
$$C = 2.1\cdot 10^{-14}\,W^{-1}, \qquad \hbar\omega_{AS} = \hbar\omega_s + (E_s - E_l),$$

(all angular frequencies are given in cm^{-1}); $\Delta\omega_{AS}$ is detuning from the intermediate level, which in the exact resonance case must be replaced by the Doppler linewidth. Three typical optical focusing configurations are here considered for the same cell of overall length L=80cm, and potassium vapor confined in the medium region length l=40cm. For calculations we consider a typical potassium atom density $N_K=10^{16}\mathrm{cm}^{-3}$ with a helium buffer gas pressure of 4 Torr (see /5/, p.154). It seems reasonable to focus the IR beam in the middle of the left subcavity at a distance 10cm from the left mirror in our case. The first process of SRS from λ_p=404nm to λ_{IR}=1.209 µm takes place in the whole discharge length L, and the second SASRS from λ_{IR} to λ_{XUV}=64nm only in the potassium filled region of length l. This part of the cell is enlightened by a cone of IR radiation with vertex angle 2θ. For example we give here three focusing cases: tight focusing with confocal parameter $b=L/40$ (α-case), medium with $b=L/4$ (β) and weak focusing with $b=L$ (γ). Here as usually $b=2\pi w_0^2/\lambda_{IR}=2\lambda_{IR}/\pi\theta^2$ where w_0 is the gaussian beam waist radius in the focus. Gain can be calculated now using eqs (2) and (3) with $\Delta\omega=0.5\mathrm{cm}^{-1}$ because we have exact resonance with intermediate level and Doppler width must be used, and $\Gamma=0.05\mathrm{cm}^{-1}$ is the Raman width (see /5/ for discussion). Then the number of XUV photons is

$$n_{XUV} = \exp(AP_{IR}) \quad \text{for} \quad n_{XUV} < n_{XUV}(\text{sat}). \qquad (4)$$

where P_{IR} is the power of $\lambda_{IR}=1.209\mu$m radiation and

$$A=\begin{cases} 6.5\ \mathrm{MW}^{-1} & \text{for } (\alpha)\\ 32\ \mathrm{MW}^{-1} & \text{for } (\beta)\\ 63\ \mathrm{MW}^{-1} & \text{for } (\gamma)\end{cases} \qquad n_{XUV}(\text{sat})=\begin{cases} 4\cdot 10^{12} & (\alpha)\\ 4\cdot 10^{11} & (\beta)\\ 2.2\cdot 10^{11} & (\gamma)\end{cases}$$

The saturated number of XUV photons here is taken equal to the overall number of storage metastable $K^{**}(3p^5\,3d4s\ ^4P^\circ_{5/2})$ atoms in the SASRS cone calculated with their density $N^{**}=8\cdot 10^{11}\mathrm{cm}^{-3}$ which is available on the present state of art of the pulsed discharge /9/. This density is very important because it is limiting the XUV output. As it can be seen from the Table the saturation input power is very near to threshold. XUV output pulse energy threshold is taken $10^{-2}\mu J$ as usually for stimulated Raman processes (/5/ p.94).

III - DOUBLE RAMAN SCATTERING SCHEME

As it was shown in the previous section IR pump power more than 0.35MW (see the Table) with line width less than $0.05\mathrm{cm}^{-1}$ is needed to achieve 64nm generation in the He-K discharge afterglow. Because the absolute values of energies of the potassium quartets are not measured with a good accuracy /15/ than tunability 100cm^{-1} of pumping radiation is needed. We analyse in this section the possibility to use the same He-K discharge for receiving this IR coherent radiation. The most convenient was found to be SRS from He 1s2p^{3}P initial level to 1s3p^{3}P final level with 1s5d^{3}D intermediate state (Fig.1). To produce λ_{IR}=1.209μm pumping p=404nm must be used, which is easily available from a powerfull and good beam quality dye-laser. Using relations (2) and (3) with detuning $\Delta\omega$=81cm^{-1}, Raman linewidth Γ=0.01cm^{-1} and oscillator strenghts f_{si}=0.0738, f_{ie}=0.0474 /16/ we have $\alpha_s = 4.5\cdot 10^{-23}$ W^{-1}cm^4. As the measured density of He(2p^{3}P) states in He-K pulsed discharge suitable pressure (4 Torr) is $N_p = 10^{12}$cm^{-1} /12/ the Stokes plane-wave gain coefficient is

$$\tilde{g}_s = \alpha_s N_p = 4.5\cdot 10^{-5}\text{cm/MW} \qquad (5)$$

To have low Stokes SRS threshold an internal spherical resonator must be used thus achieving high conversion efficiency /13,14,17/. The losses are small because good quality mirrors are available for the near IR. The number of passes in the active medium is $n = c\tau/L$, where τ is the pumping pulse duration which must be less than 100ns which is duration of population inversion in the afterglow of He-K discharge. The maximum possible conversion efficiency (relation of the IR output power to the p=404nm input power: $\eta = P_{IR}/P_p$) is $\eta = (\omega_s/\omega_p)R^{n-1}$, where R is the mirror reflectivity. In our calculations reflectivity is taken egual to 99%. For a focused Gaussian beam, gain per pass G_1 is in fact not a function of pump intensity but of pump power P_p and is given by /13,17/:

$$G_1 = \frac{4P_p\tilde{g}_s}{\lambda_p + \lambda_s}\, tg^{-1}\left(\frac{L}{b}\right) \qquad (6)$$

were g_s is given by (5) and b is the confocal parameter. The overall gain coefficient for n passes is

$$G_n = \frac{1-R^n}{1-R}\, G_1 + (n-1)\ln R. \qquad (7)$$

Conversion efficiency versus the number of passes, with pump depletion and mirror reflectivity taken into account /17/ can be written in a simple form

$$\eta_n = \frac{\eta_m}{1 + \frac{\eta_m}{r}\, e^{-G_n}} \qquad (8)$$

where r is the ratio of spontaneous Stokes noise power P_{so} to the input pump power P_p, $P_{sc} \sim h\nu_s\Delta\nu_s$. With a Stokes Raman linewidth of the same value as used before (0.01cm^{-1}) $P_{so} \sim 5.10^{-9}$W and for an input power $\sim$5MW we have $r \sim 10^{-16}$. Calculating G_n from (7) and η_n from (8) we obtain $P_s = \eta_n P_p$ as a function of n or in other words as a function of the pumping pulse duration. Here rectangular pump pulse is considered with slopes less than half duration of one pass in the resonator, which is equal to 2.7ns. Then using (4) we can obtain anti-Stokes SRS gain and XUV output energy $W_{XUV} = h\nu_{XUV}\, n_{XUV}$. This energy for the three focusing cases we are studying is given in Fig.2 as a function of the dye-laser input power. In the Table main parameters of the three focusing configurations are summarized. The α-case has the lowest power threshold for travelling-wave excitation because it is the case of tight focusing. But for intraresonator enhanced anti-Stokes wave the lowest threshold is for the γ-case (only 1.5MW). Also the saturation is reached earlier for γ, but the output energy value is the lowest due to the small pumping cone volume. In the tight focusing case α, the output is high enough up to 12.3μJ per pulse. Also the highest efficiency: 2.10^{-4} without resonator may be achieved, but in this case the input power must be very high ~ 22MW. It can be reduced by intraresonator SRS. For L=80cm resonator length the optimum pulse duration is calculated to be 5.4ns, and pump power reduces to 13.8MW; efficiency is then 1.7×10^{-4}. The shortcoming of tight focusing is high pump-power intensity in the focus which can lead to some unwished effects. This intensity is a minimum in the γ-case. Pumping power (λ_p=404nm) as a function of pumping pulse duration is given in Fig.3. It may be seen that all the curves have minima that means optimum pumping pulse duration.

IV - SUMMARY

In this paper the anti-Stokes Raman laser based on collisional energy transfer from $He(2s^3S)$ metastable atoms excited in a discharge afterglow to quartet metastable $3p^53d4s\ ^4P_{5/2}$ state of potassium is analysed for different pumping beam focusing parameters. A new method is proposed to produce the needed IR radiation for the anti-Stokes operation in the same discharge medium, by stimulated SRS of a convenient visible dye-laser light, also using IR resonator for this Stokes radiation. Principal results are shown in the Table and several conclusions can be drawn:

1. The lowest threshold for XUV λ_{AS}=64nm generation may be achieved by using 0.35MW radiation at λ =1.209μm by focusing it into the potassium vapor column (l=40cm) with storage $3d^5\ 3d4s^4\ P_{5/2}$ atoms concentration $N^{**} \approx 8.10^{11}cm^{-3}$ and long focusing (b=80cm, γ-focusing case).
2. The maximum XUV output is limited by saturation due to the limited number of quartet K** metastables inside the pumped volume. This number may be rised, increasing K** concentration by optimizing discharge conditions (electric power, duration, etc.) or by increasing the pumped volume.
3. λ_p=404nm pump radiation may be used to produce the needed IR radiation in the same cell by Stokes SRS, from the He $3p\ ^3P$ levels excitet in the same discharge. Large overall conversion efficiency and high output (12μJ) may be achieved in the tight focusing case (α). But the pump intensity in the focus is very high in this case. On the contrary for along confocal parameter (γ-case) pump intensity in the focal plane is low.
4. To reduce the dye-laser pump power an IR resonator may be used. Because high quality mirrors in this wavelength region are available, than multipass gain may be achieved. Thus the 404nm pump threshold may be lowered down to 1.5MW for 8ns pulse radiation in the γ-case. Maximum XUV output, equal to 12.3μJ , is reached for a 3.8MW, 5.4ns dye-laser pumping pulse in the α-case, with overall efficiency 1.7×10^{-4}.

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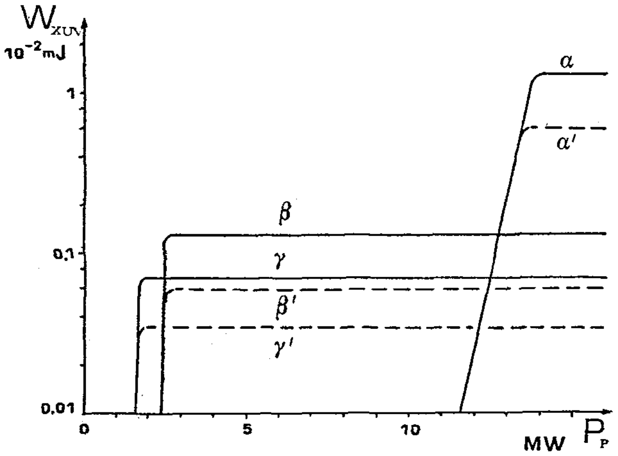

Fig.2 - Output pulse energy at λ =64nm as a function of pumping violet λ_p=404nm power with IR resonator used and pumping pulse duration 8ns for α , 40ns for β and 48ns for γ-case; an initial quartet metastable potassium atom density of $8 \cdot 10^{11} cm^{-3}$ (full curves) or $4 \cdot 10^{11} cm^{-3}$ (dotted curves and primed letters).

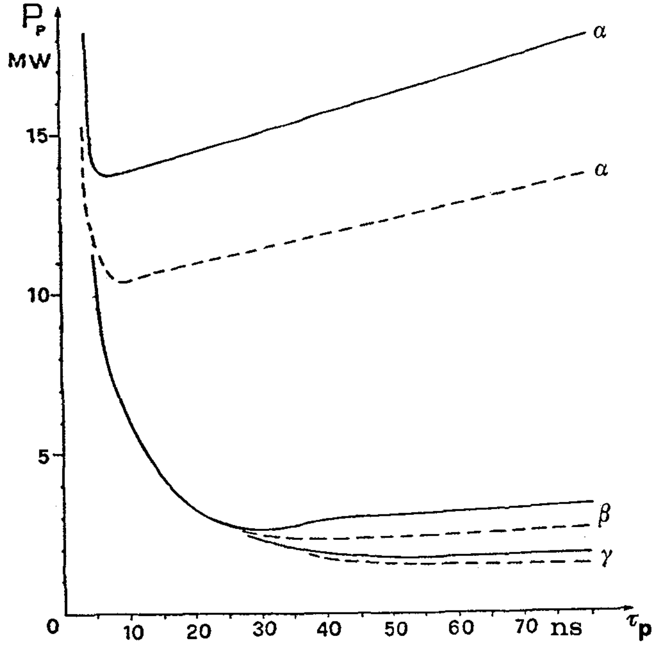

Fig.3 - Pumping power of λ_p =404nm pulse needed for achieving XUV generation threshold (dotted lines) and saturation (full lines) versus pumping pulse duration. α ,β β and γ are for corresponding focusing parameters.

TABLE.XUV GENERATION PARAMETERS

Parameter			Unit	Focusing cases		
				α	β	γ
Confocal parameter (b)			cm	2	20	80
XUV beam divergence (2θ)			mrad	13	4	2
K** atoms pumped optically for a density $N^{**}=8\cdot10^{11}\,cm^{-3}$				$4\cdot10^{12}$	$4\cdot10^{11}$	$2.2\cdot10^{11}$
XUV threshold (pulse energy 10^{-2} μJ)	Minimum density N**		cm^{-3}	$6.5\cdot10^{8}$	$6.5\cdot10^{9}$	$1.2\cdot10^{10}$
	Minimum IR power		MW	3.4	0.68	0.35
	Minimum λ_p =404nm pump power	Travelling wave	MW	20.8	22.6	36.7
		Resonator		10.5	2.3	1.5
	Pulse duration for min. power		ns	8	40	48
Maximum XUV pulse energy (at saturation)			μJ	1.23	0.12	0.07
Max. XUV pulse energy (saturation)	Minimum IR power needed for saturation		MW	4.5	0.83	0.42
	Minimum λ_p =404nm pump power	Travelling wave	MW	21.3	22.7	37.0
		Resonator		13.8	2.5	1.7
	Pulse duration for min. power		ns	5.4	32	40
	overall maximum conversion efficiency	Travelling wave (pulse duration 2.7ns)	10^{-4}	2.1	0.2	0.07
		Resonator		1.7	0.2	0.12
	Pump beam intensity in the focal plane	Travelling wave	$\frac{GW}{cm^2}$	176	18.8	7.7
		Resonator		114	2.1	0.36