Appl. Phys. B 34, 171–173 (1984)

Applied Physics B Photophysics and Laser Chemistry

Ninth-Order Nonlinear Polarization and vuv Generation in Hg Vapor

V. F. Lukinykh, S. A. Myslivets, A. K. Popov, and V. V. Slabko

L. V. Kirensky Institute of Physics, USSR Academy of Sciences, Siberian Branch, SU-660036 Krasnoyarsk, USSR and Krasnoyarsk State University

Received 22 November 1983/Accepted 25 January 1984

Abstract. Interference of resonant ninth-order and nonresonant seventh-order nonlinear polarizations in Hg atoms has been studied, and direct conversion of fundamental ir radiation into the vuv has been obtained. The interference has been observed in the form of a Fano-Boitler contour. The efficiency of generation by ninth-order processes appears to be higher than that by seventh-order.

PACS: 42.65, 42.80

Second-order nonlinearities of crystals and third-order nonlinearities of free atoms and molecules are generally used in nonlinear optics. Frequency mixing in gases and metal vapors seems to be the most promissing technique to produce tunable coherent vacuum ultraviolet (vuv) and soft x-ray radiations. The study of higher-order nonlinearities is of a particular interest due to the possibilities of producing radiation in the extreme ultraviolet [1–3]. This paper reports on the study of peculiarities of resonant and nonresonant higher-order nonlinear processes in Hg vapor in a strong pump field. The possibility is shown to directly convert ir radiation to the vuv with an output power of the order of 0.1 kW.

1. Frequency-Mixing Schematic

Nonlinear polarization is strongly enhanced with the pump frequency combinations coming close to the frequencies of atomic transitions. This is also followed by an increase of atomic perturbation and results in limiting linear and nonlinear competing processes. Earlier we have employed third-order nonlinearity of Hg atoms for resonant tripling of fourth-harmonic of Nd:glass laser radiation to generate the twelfth harmonic in the range of 89.6 nm [4]. Four-wave mixing in Hg vapor is coming into still wider use to produce tunable vuv radiation [5–8]. Under certain conditions higher-order resonant nonlinear polarizations can exceed lower-order nonresonant ones. In the present paper we demonstrate this for Hg vapor and employ it for direct conversion of fundamental ir radiation to the vuv. The higher-order processes have been selected to obtain the vuv radiation for which some optical materials are still transparent and hence a closed vapor-cell with windows can be utilized. Seventh-harmonic generation ($\lambda = 153.6$ nm) of frequency tunable Nd:glass laser radiation both by ninth-($\omega_s = 8\omega - \omega$) and seventh-($\omega_s = 7\omega$) order processes is investigated. The frequency 8ω can be tuned over the same resonance $6^1S_0 - 8^1S_0 (74{,}404\,\mathrm{cm}^{-1})$ as in [4]. Frequency-degenerate, higher- and lower-order processes interfere. The ninth-order process can be discriminated due to its resonant properties. When tuning the fundamental frequency over an eight-photon resonance, the real part of the resonant nonlinear susceptibility changes its sign. That is why frequency dependence of the output power would acquire the characteristic shape of a Fano-Boitler resonance, having been studied earlier for nonlinear processes involving transitions via the continuum [9, 10].

2. Experimental

The input radiation was supplied by a Nd:glass laser system. The system consisted of an actively mode-locked oscillator tunable in the range of 1052.5–1078.9 nm, a spatial filter and a double-stage

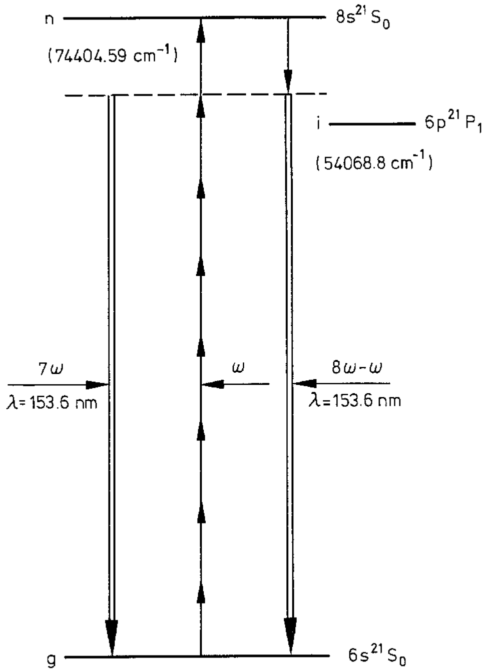

Fig. 1. Energy level diagram of Hg atoms giving the main contribution to the upconversion process

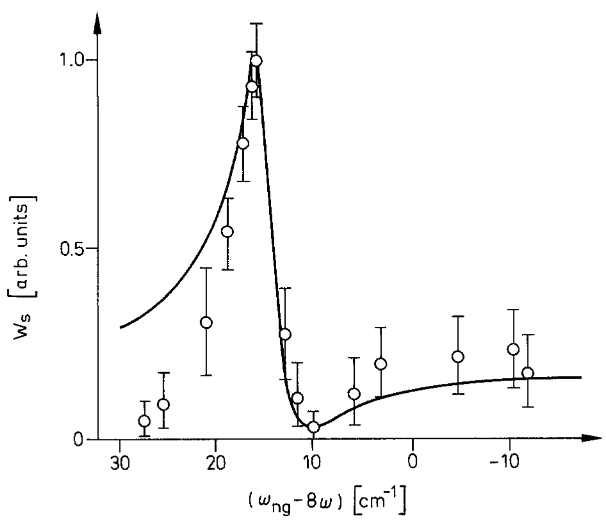

Fig. 2. Dependence of the seventh-harmonic power of Nd : glass laser radiation (arbitrary units) on eight-photon resonance $6s^{21}S_0$—$8s^{21}S_0$ detuning

travelling wave amplifier. A 10-cm focal lens focused the pump radiation into the center of an about 5-cm long stainless-steel heat-pipe cell with Hg vapors. The confocal parameter was about 2 cm. The cell was attached to the front of a vacuum monochromator VMR-2. A heated LiF window separated volumes of the cell and the monochromator. A secondary multiplier VEU-4 was used as a photodetector.
The arrangement ensured control over the frequency, spectral width, energy and temporal shape of the input signal. Under optimum conditions the latter was observed as a 100 ns train of 14 ns-separated pulses with 4×10^{-10} s duration. In the wavelength range of 1075.2 nm, corresponding to an eight-photon resonance the average pulse intensity in the focus yielded a magnitude of the order of 10^{12} W/cm^2. The spectral width of the first-harmonic signal was 2 cm^{-1}.

3. Theoretical Discussion and Experimental Results

The output power W_7 at the frequency 7ω is proportional to the squared modul of nonlinear polarization P^{NL}:

$$P^{\mathrm{NL}}=P^{(7)}_{(7\omega)}+P^{(9)}_{(7\omega=8\omega-\omega)}=\chi^{(7)}_{(7\omega)}E^7+\chi^{(9)}_{(7\omega=8\omega-\omega)}E^7|E|^2 \quad , \tag{1}$$

where E is the complex amplitude of the incident radiation, $\chi^{(7)}_{(7\omega)}$ and $\chi^{(9)}_{(8\omega-\omega)}$ refer to the seventh- and ninth-order nonlinear susceptibilities, respectively,

$$\chi^{(9)}_{(8\omega-\omega)}=\chi^{(7)}_{(7\omega)}\cdot\hbar^{-2}(\omega_{ng}-8\omega+\delta+\mathrm{i}\,\gamma_{ng})^{-1}\sum_{i,j}\frac{d_{in}d_{nj}}{(\omega_{jg}-7\omega)} \quad . \tag{2}$$

ω_{ng}, ω_{jg} and ω are the frequencies of $6^1S_0-8^1S_0$ transition, intermediate transitions and of the incident radiation, respectively; g is the ground state; d_{in} and d_{nj} denote electric dipole momenta of the transitions; γ_{ng} is $(n-g)$ transition halfwidth accounting for power-broadening and radiation spectral width; δ is the ac Stark shift of the $6^1S_0-8^1S_0$ transition in a pump field. Then, disregarding the inhomogeneity of the pump field we have

$$W_7\sim|P^{\mathrm{NL}}|^2=|\chi^{(7)}_{(7\omega)}|^2\left(1+\frac{A^2+2Ax}{1+x^2}\right)(|E|^2)^7 , \tag{3}$$

where $x=(\omega_{ng}-8\omega+\delta)/\gamma_{ng}$ is a normalized detuning; and

$$A=\frac{|E|^2}{\gamma_{ng}\hbar^2}\sum_{i,j}\frac{d_{in}d_{nj}}{(\omega_{jg}-7\omega)} .$$

The frequency dependence of seventh-harmonic power is shown in Fig. 2. The points are the experimental values of W_7. A theoretical curve has been constructed according to (3) with $A=1.92$. The value of $x=0$ is 16 cm^{-1} shifted from the nonperturbed eight-photon resonance which agrees with the estimated magnitude of the ac Stark shift $\delta\approx10$ cm^{-1} in the field of 1×10^{12} W/cm^2 intensity. The maximum conversion efficiency corresponding to eight-photon resonance is 5 times higher than that at a considerable detuning from the resonance when the major contribution is made by the seventh-order process

Equation (3) shows the curve in Fig. 2 to be determined by A parameter. Assuming the main contribution to A to be given by the $6^1P_1 - 8^1S_0$ transition, its oscillator strength can roughly be estimated. The order of the calculated value of $f_{6^1P_1-8^1S_0} = 0.026$ coincides with that of $f = 0.043$ from [11] which supports the main features of the model interpreting the experimental dependence.

The conversion efficiency being mainly determined by the ninth-order process reached 3×10^{-6} which corresponds to $\sim 10^{13}$ photon/pulse and the output power of about 0.1 kW. These values are comparable to the typical conversion efficiencies obtained by third-order processes and are among the highest available for higher-order nonlinearities [12].

In the presented conversion scheme the seventh-harmonic radiation can also result from lower-order processes both nonresonant ($3\omega + 3\omega + \omega = 7\omega$, $5\omega + \omega + \omega = 7\omega$) and resonant ($5\omega + 3\omega - \omega = 7\omega$; $3\omega + 3\omega + \omega + \omega - \omega = 7\omega$), the third- and fifth-harmonics being generated in the same nonlinear medium. Estimations, however, show that the cascade processes are rather strong at concentrations of $N > 10^{22}\,\mathrm{cm}^{-3}$ [13]. Hence for our case at $N \sim 4 \times 10^{16}\,\mathrm{cm}^{-3}$ these do not contribute sufficiently to the conversion process. Assuming a resonance width of $\gamma_{ng} \sim 2\,\mathrm{cm}^{-1}$ for the conversion scheme under consideration estimates yield $\chi^{(7)}/N \sim 10^{-59}$ cgse units for the nonresonant susceptibility and $\chi^{(9)}/N \sim 10^{-68}$ cgse units for the resonant one.

The above expressions are valid for a homogeneous plane wave in the absence of a phase-mismatch ($\Delta k = 0$). The experimental pump radiation corresponds to a Gaussian mode with the confocal parameter $b \approx 2$ cm, the active zone being 5 cm long. The analysis of phase-matching conditions for tight focusing shows different values of optimum vapor concentrations for the seventh- and ninth-order processes. This results in the concentration dependence of the resonance shape (parameter A), which was observed experimentally. Moreover, the ac Stark shift is as well a function of time and of spatial coordinates. This should bring about an effective broadening of the resonance mentioned. These aspects however need a more detailed treatment.

Acknowledgements. The authors are grateful to Yu. I. Heller for helpful discussions and to R. Wallenstein for discussions and for the copy of his paper [6] prior to its publication.

References

1. S.A. Akhmanov: In *Nonlinear Spectroscopy*, ed. by N. Bloembergen (Mir, Moscow 1979) pp. 323–326
2. J. Reintjes: Appl. Opt. **19**, 3889 (1980)
3. J. Bokor, P.H. Bucksbaum, R.R. Freeman: Opt. Lett. **8**, 217 (1983)
4. V.V. Slabko, A.K. Popov, V.F. Lukinykh: Appl. Phys. **15**, 239 (1977)
5. K.S. Hsu, A.H. Kung, L.J. Sich, J.F. Young, S.E. Harris: IEEE J. QE-**12**, 60 (1976)
6. R. Hilbig, R. Wallenstein: IEEE J. QE-**19** (1983)
7. R. Wallenstein: Laser Optoelektronik **3**, 29 (1982)
8. R. Mahon, R.C. Tomkins: IEEE J. QE-**18**, 913 (1982)
9. Yu. I. Heller, A.K. Popov: *Laser-Induced Nonlinear Resonances in the Continua* (Nauka, Novosibirsk 1981)
10. S.S. Dimov, L.L. Pavlov, K.V. Stamenov, Yu. I. Heller, A.K. Popov: Appl. Phys. B **30**, 35 (1983)
11. G.L. Kasabov, V.V. Eliseev: *Spectroscopic Tables for Low-Temperature Plasma* (Atomizdat, Moscow 1973)
12. D.C. Hanna, M.A. Yuratich, D. Cotter: *Nonlinear Optics of Free Atoms and Molecules* (Springer, Berlin, Heidelberg, New York 1979) p. 351
13. J. Reintjes, C.Y. She, R.C. Eckardt: IEEE J. QE-**14**, 581 (1978)

Responsible for Advertisements: E. Lückermann, G. Probst, Kurfürstendamm 237, D-1000 Berlin 15, Telephone: (0 30) 8 82 10 31, Telex 01-85411
Responsible for the Text: H. K. V. Lotsch, Tiergartenstrasse 17, D-6900 Heidelberg. Printers: Brühlsche Universitätsdruckerei, Giessen, Printed in Germany