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Continuous-Wave Frequency Mixing and UV Generation in Sodium Vapor

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Abstract. Resonant nonlinear four-wave mixing processes have been studied in sodium vapor. The generation of cw uv radiation and the upconversion of $\lambda = 10.8\,\mu m$ light is reported. The coefficient $C = P_4/P_1P_2P_3$ obtained was on the order of $\sim 10^{-2}\,W^{-2}$, the pump spectral width being 12 GHz. Resonant atomic nonlinearities are shown to be used for effective cw frequency conversion.

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CW nonlinear optical processes are of great practical and scientific interest, providing the best resolution in nonlinear spectroscopy and the highest energy contribution in the selective phenomena. The drawback, however, is that the intensities of cw output laser fields are generally far less compared with the pulse ones, which makes these processes difficult to utilize, especially to extend the frequency conversion to the short-wavelength cw ranges.

Low field intensity could be compensated for by resonant enhancement of the nonlinear susceptibility. Then, however, the competing processes resulting in perturbation of the absorption, nonlinear susceptibilities and etc. would become important. The problem of maximum conversion efficiency and the optimum conditions to realize it in weak cw fields still remains less investigated and needs a further detailed treatment. The lack of theory can be explained by the great number of the factors to be taken into account. That is why obtaining and classifying the experimental data is of importance.

In recent years much attention has been paid to four-wave frequency mixing in gaseous media, most of the works being devoted to the frequency conversion of pulsed laser radiation and only a few of them dealing with cw light. In four-wave frequency mixing the output power may be represented as $P_4 = CP_1P_2P_3$, P_1, P_2, and P_3 being the pump powers, and C the coefficient proportional to the squared third-order nonlinear susceptibility of the medium. Frequency tunable cw dye lasers produce 4–5 orders lower power than the pulsed ones and hence allow 12–15 orders lower output in four-wave mixing. That is why even the possibility to detect these processes in cw operation seemed problematic at first.

For that reason, in [1] the effect of atomic velocity distribution and intermediate-level population on completely resonant interaction was studied in four-level schemes. References 2, 3 report theoretical and experimental investigations of quasidegenerate four-wave mixing $\nu_4 = \nu_1 + \nu_1 - \nu_2$, the interaction frequencies being resonant within the natural linewidth. Two-, three-, and four-level schemes were realized between the sublevels of a degenerate two-level system. The output signals have been detected and used for spectroscopic purposes with 10^{-2} W He-Ne laser fields as the input waves. Resonant sum-frequency mixing in Ne was achieved in [4] inside the cavity of a He-Ne laser operating at several transitions simultaneously. In [5] two-photon resonant four-wave mixing has been obtained in Sr by summing the frequencies of argon-ion and dye lasers; it was used to produce cw coherent vuv in the region of 170 nm. A three-photon resonance with the autoionizing level was provided by applying an axial magnetic field (50 kG) to the nonlinear medium. Recently two-photon resonant single-frequency third-harmonic generation of a cw dye laser has been reported in Mg [6]. The vuv intensity at

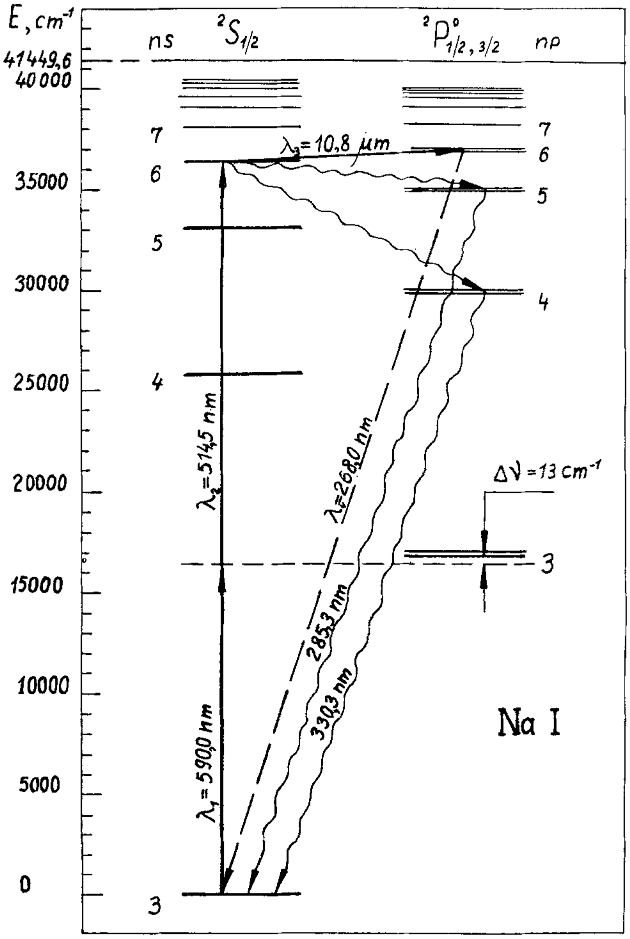

Fig. 1. Partial energy diagram of S and P sodium levels

$\lambda = 143.6$ nm was of the order of 10^5 photon/s. This value is quite sufficient for spectroscopy taking into account that the spectral width of the vuv output is less than 1 MHz.

The present paper studies resonant four-wave mixing $\nu_4 = \nu_1 + \nu_2 + \nu_3$ in sodium vapor. Under exact two-photon resonance and small detunings from the intermediate one- and three-photon resonances a cw coherent uv radiation with $\lambda_4 = 268$ nm has been generated. CO_2-laser radiation has been upconverted to this wave range. Resonant atomic nonlinearities are shown to be utilized for a fairly efficient frequency conversion and for developing cw short-wavelength sources.

1. Experimental

Figure 1 shows a partial energy-level diagram of a sodium atom and the frequency conversion scheme for radiation with $\lambda_1 = 590$ nm, $\lambda_2 = 514$ nm, and $\lambda_3 = 10.8$ μm. The radiation at λ_1 and λ_2 are two-photon resonant. The detuning from the intermediate level $3P_{1/2}$ is 13 cm^{-1} and the sum-frequency $\nu_4 = \nu_1 + \nu_2 + \nu_3$ is 0.14 cm^{-1} detuned from the frequency of $3S - 6P_{3/2}$ transition.

The three incident cw fields are: Ar^+-laser-excited dye-laser radiation with a spectral width of 12 GHz, a green line of a single-mode Ar^+-laser with the spectral width of 5 GHz and the line-tunable single-frequency CO_2-laser radiation. These were focused ($f = 15$ cm) into the cell with Na vapors. The output at the sum-frequency was preselected by a uv filter and then selected by a difraction monochromator. To improve the sensitivity of the detection, a modulation technique was employed. This is based on modulating the amplitude of one of the pump waves and selecting the signal by a detector and an amplifier tuned to the modulation frequency.

Sodium vapor was produced in a stainless steel heatpipe oven [7] (20 cm long with an inner diameter of 16 mm). To prevent the windows from Na condensation the cell contained helium.

Fine tuning to the two-photon resonance was ensured by monitoring the spontaneous radiation. The monochromator was tuned to one of the spontaneous emission lines (wavy arrows in Fig. 1) resulting from a two-photon excitation of Na atoms to $6S$ state and a subsequent spontaneous decay. The uv radiation from the cell was analyzed both in the absence and in the presence of CO_2-laser light.

In the absence of a CO_2-laser beam the uv spectrum consists of two strong lines with $\lambda = 285.3$ and 330.2 nm and a weak one with $\lambda = 268$ nm (Fig. 2a). The intensity of the $\lambda = 268$ nm line displays a sharp increase at switching on 10.8 μm radiation while for the other two lines it decreases by 7–8% (Fig. 2b). The appearance of the lines has been found to be closely correlated with the two-photon excitation of the $6S$ state of Na atoms. When any of the pumps providing the two-photon excitation is absent, no radiation has been observed at these uv lines, whereas if both waves are present, the uv intensities drastically fall when the detuning from the two-photon resonance is of the order of the radiation's spectral width. The uv radiation has a wide angular divergence except the one at $\lambda = 268$ nm. This radiation is directional with the divergence close to that of the pump waves ($\sim 10^{-2}$ rad) and the power depends linearly on the power of the CO_2-laser. The lines at $\lambda = 285.3$ and 330.2 nm are associated with the radiative decay of the $6S$ state via the intermediate levels $5P$ and $4P$, respectively. The radiation at $\lambda = 268$ nm having a small angular divergence which appears in the presence of all the input beams, can be ascribed to the four-wave mixing.

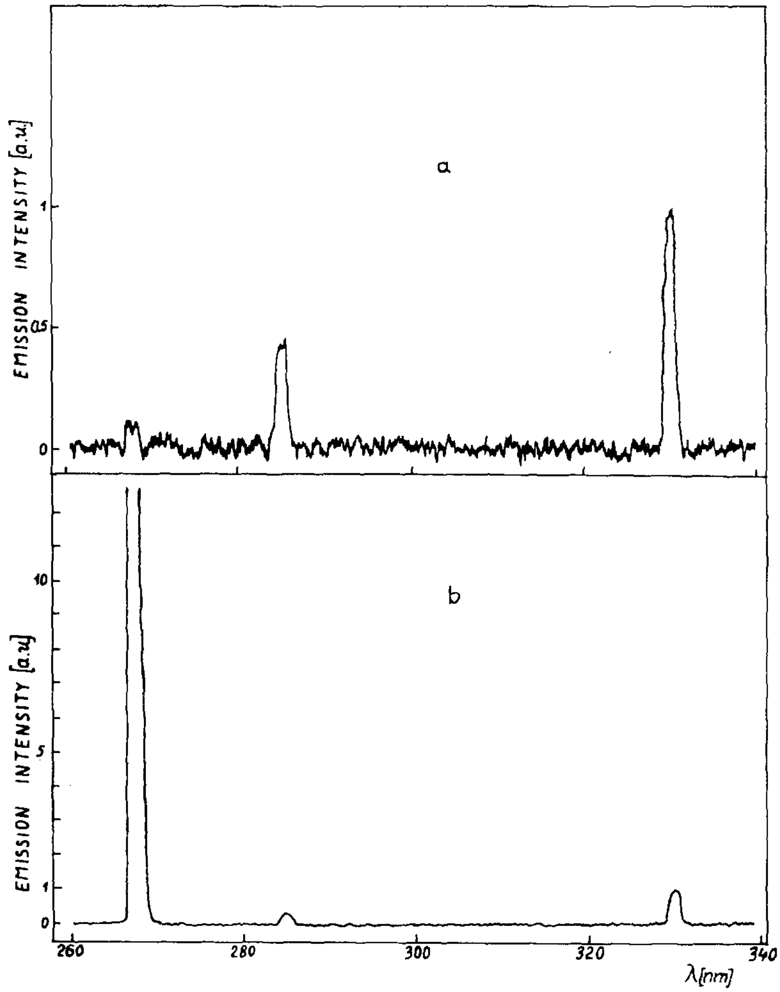

Fig. 2a and b. Radiation spectra of sodium vapor: (a) for the two-photon transition $3S$–$6S$ excited by radiation with $\lambda_1 = 590$ nm and $\lambda_2 = 514.5$ nm; (b) for the interaction with the three pump waves: $\lambda_1 = 590$ nm, $\lambda_2 = 514.5$ nm, and $\lambda_3 = 10.8$ μm

Weak radiation at the $6P$-$3S$ transition frequency close to the sum frequency $\nu_4 = \nu_1 + \nu_2 + \nu_3$ in the absence of the pump wave at ν_3 may result from a number of processes. Their analysis is beyond the scope of the present paper.

Two-photon resonant four-wave mixing was observed for the wavelength of CO_2-laser radiation tunable in the 10.4–11.1 μm interval with the sum-frequency output maximized for $\lambda_{ir} = 10.8$ μm. It is this line that corresponds to the minimum three-photon detuning $\Delta\nu = -0.14\,cm^{-1}$. At the pump powers $P_1 = 40$ mW, $P_2 = 700$ mW and at the ir power passing through the pump-beam waist, $P_3 = 0.5$ mW, the highest sum-frequency output yielded a magnitude of the order of 10^{-7} W which makes up 10^{11} photon/s. Thus, the proportionality coefficient C was measured to be $C \sim 10^{-2}\,W^{-2}$ and the power conversion efficiency was about 2×10^{-4}. These values are encouraging taking into account that the conversion efficiency is inversely proportional to the pump's spectral bandwidth. Hence they can be magnified by its narrowing, by increasing the pump powers as well as by improving the beam's spatial matching.

2. Discussion

For cw vuv generation in Sr vapors [5] the coefficient obtained was of the order of $10^{-10} - 10^{-11}\,W^{-2}$. It is likely to be due to smaller values of the dipole transition moments, substantial one-photon detuning ($700\,cm^{-1}$) from the intermediate level at the two-photon transition and the large width of the autoionizing resonance. The same value $C \sim 10^{-11}\,W^{-2}$ has been achieved in [6]. For the comparison with our work, the coefficient $C \sim 10^{-7}\,W^{-2}$ follows from the results of [8], where a similar frequency-mixing scheme was studied for Na vapor in the pulse regime with the only difference in the one-photon detuning from the intermediate level $3P$ which was considerably higher ($600\,cm^{-1}$). The nonlinear susceptibility is inversely proportional to this detuning and the coefficient C – to the square of the ratio: atomic density/detuning. Thus the orders of magnitude of the coefficient C estimated for similar densities and detunings from the results of [8] and reported here are in a fair agreement. Estimates derived from direct calculation and from the experimental results give, for the pumps used, the atomic nonlinear susceptibility magnitude of $|\chi^{(3)}| \sim 10^{-24} - 10^{-25}$ esu. This, when compared to $|\chi^{(3)}| \sim 10^{-31}$ esu estimated in [6], supports the suggestion [6] of the most efficient cw four-wave mixing in metal vapors to be realized due to the tunable three-frequency pump and the corresponding enhancement of the atomic nonlinear susceptibility by a proper choice of one- and three-photon detunings. At present the maximum coefficient C values, as far as we know, have been obtained under exact resonances over all one- and multiphoton transitions at the level population densities of the order of about 10^{-9} cm^{-3}. They yield $\sim 10^{-2}$ W^{-2} [2] and $\sim 10^{-3}$ W^{-2} [4].

Thus the results obtained have proved the nonlinear resonant processes under optimum conditions to be capable of extending cw laser sources to new spectral ranges and of improving nonlinear spectroscopy techniques. They have also given an impetus to the development of the corresponding theory.

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Note added in proof: At $P_1 = 0.13$ W, $P_2 = 0.33$ W, $P_3 = 0.3$ mW and improved focusing conditions the output reached $P_s = 10^{-5}$ W and hence $\eta_p = 3\%$, $C \approx 0.8$ W^{-2}.

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