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Resonant Generation of Even-Order Harmonics in Metal Vapors

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Abstract. Generation of the second- and fourth-order harmonics of laser radiation is reported in Mg vapor at the forbidden $3s^{2\,1}S_0 - 4s^1S_0$ transition. The conversion efficiency for the second-order harmonics has been measured to be $\sim 10^{-2}\%$ and for the fourth one $\sim 10^{-8}\%$.

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When studying nonlinear interactions in gaseous media the major contribution is assumed to be given by odd-order nonlinearities, beginning with the third. However, under certain conditions even-order nonlinearities become allowed in isotropic media. These are the ones either destroying the central symmetry of the medium or involving high-order multipole transitions – magnetodipole and electric quadrupole.

Under nonresonant conditions symmetry breaking occurs at high pump intensities [1–3], though moderate intensities are sufficient when an even number of pump photons is resonant to some forbidden medium transition [4–8]. Higher multipole momenta enter into the process when the use is made of either external static electric or magnetic fields or of a special noncollinear geometry of the pump beams [9–11].

Of particular interest is the generation of even-order harmonics in alkaline-earths via the transitions of the type $ms^{2\,1}S_0 - ns^1S_0$. The process happens in spite of the transitions being electrodipole-, electroquadrupole-, and magnetic dipole-forbidden and does not require the application of both external field and a special interaction geometry.

Earlier SHG of dye-laser radiation has been reported in Ba vapor at the transition $6s^{2\,1}S_0 - 6p^{2\,1}S_0$ [5], in Ca vapor at the transition $4s^{2\,1}S_0 - 5s^1S_0$ [6], and in Zn vapor at the transition $4s^{2\,1}S_0 - 5s^1S_0$ [7].

In this paper we are reporting on the second- and fourth-harmonic generation in Mg vapor. The input frequency-tunable radiation was produced by either a N_2-laser- or ruby-laser-pumped dye laser. This was focused into a cell with Mg vapor and He as a buffer gas. The output was filtered and analyzed with a monochromator and detected by a photomultiplier and an oscilloscope. The output power was controlled by means of another photomultiplier and a second oscilloscope channel.

A SHG in Mg vapor has been observed when 20 kW radiation at 459.7 nm of Coumarine 120 in ethanol

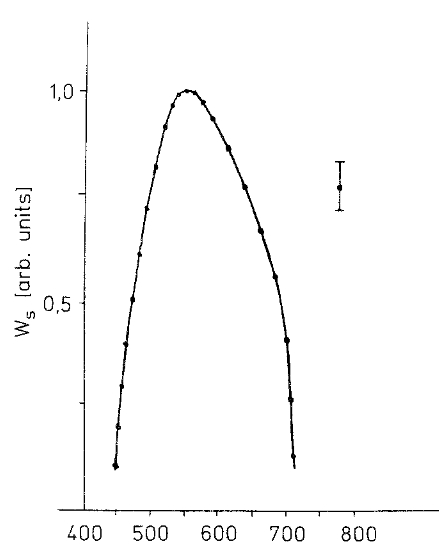

Fig. 1. Power dependence of SHG on the temperature of Mg vapor

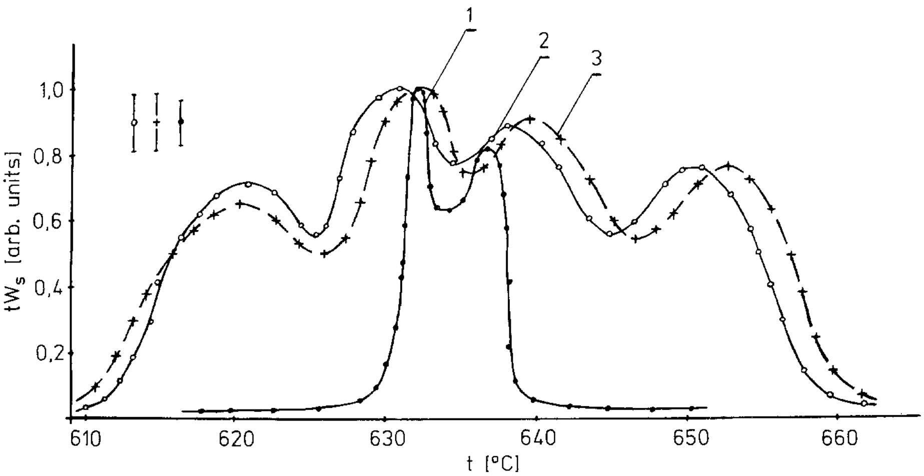

Fig. 2. Power dependence of the fourth-harmonic generation on the temperature of Mg vapor (*1* pump power: 300 kW, He pressure: 20 Torr; *2* pump power: 1.2 MW, He pressure: 20 Torr; *3* pump power: 1.2 MW, He pressure: 300 Torr)

dye-laser pumped by 7 ns pulses of N_2-laser was tuned to the two-photon resonance with the forbidden $3s^{21}S_0-4s^1S_0$ transition of Mg atom. Fourth-harmonic generation has been observed when 1.2 MW radiation at 919.5 nm of a dye-laser pumped by 30 ns pulses of a ruby laser was tuned to the 4-photon resonance with the same $3s^{21}S_0-4s^1S_0$ transition. For both cases the output wavelength was 229.9 nm. Spatial characteristics of the output and its power dependence on Mg atomic concentration manifested the parametric origin of the generation. Thus, for instance, the output at the double frequency (Fig. 1) had a typical phase matching behaviour similar to the one observed earlier [6] for Ca vapor.

The fourth-harmonic generation exhibited a different phase-matching behaviour, with several peaks depending on the pump power: for 300 kW the experimental curve showed two peaks (Fig. 2, Curve *1*) and for 1.2 MW – four peaks (Fig. 2, Curve *2*). Another peculiarity is that the fourth-harmonic generation occurs in a much shorter concentration interval than SHG, and the interval increases with the pump power. Measurements for different helium pressures showed no appreciable contribution of the buffer gas to the process (Fig. 2, Curve *3*).

The power dependence on the frequency detuning from the transition demonstrated a strongly resonant behavior, the resonant width being determined by the pump spectral width. The conversion efficiency for SHG was as high as 0.1–0.01% and for the fourth-harmonic – 10^{-6}–10^{-8}%. Similar phenomena have been observed in Sr vapor.

The experimental studies have revealed some processes breaking the symmetry of the gaseous medium and making the even-order nonlinearities allowed. This may, for example, be explained by a specific multiphoton-ionization-induced spatial charge distribution and the appearance of the electric field in the interaction volume [3]. The experimental dependences observed, however, need a more detailed theoretical treatment.

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