Six-and eight-photon resonance processes in sodium vapor

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We have made the first study of four-and six-photon resonances in atomic sodium by the methods of optical mixing spectroscopy. We have experimentally demonstrated the possibility of spectroscopy probing many-photon transitions in atoms by means of higher-order parametric effects.

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1. In this letter we present the results of experiments in which four- and six-photon resonances in sodium vapor were observed for the first time by the methods of optical mixing spectroscopy. We measured the matrix elements of the four-photon resonance $3s$–$4s$ in atomic sodium. It was shown that optical mixing spectroscopy permits doing such studies at moderate power levels of the exciting radiation. This opens up wide possibilities for investigating steady-state and transient effects at many-photon resonances.

2. Optical frequency-mixing spectroscopy is currently in wide use. Various modifications of this method have been successfully applied in the study of Raman and two-photon resonances,[1,2] polariton excitations in crystals,[3] etc. The use of picosecond techniques permits direct investigation of the dephasing time of quantum transitions in media.[1,2,4]

However, up until now all of the measurements of this type have been based on observation of the four-photon processes occurring in the third-order nonlinear coefficient $\chi^{(3)}$. It is of considerable interest to develop analogous methods based on resonant parametric processes of higher order, which would substantially extend the capabilities and applications of nonlinear spectroscopy.[5,6] No experimental research on processes of this kind had previously been done.

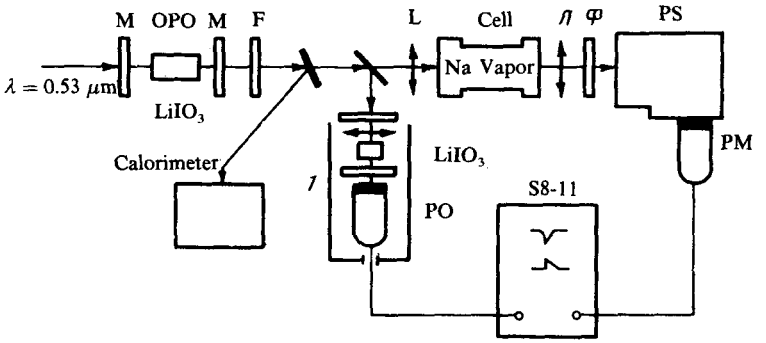

FIG. 1. Layout of main part of experimental apparatus: 1—reference channel, M—mirrors, F—light filters, L—lenses, OPO—optical parametric oscillator, PM—photomultiplier, S8-11—oscilloscope, PS—probe systems

3. In our experiments we probed the many-photon resonances by means of the simplest frequency-degenerate parametric processes of higher order—the generation of the fifth harmonic by the fifth- and seventh-order nonlinear coefficients. We chose sodium vapor as the nonlinear medium. The pump source was a tunable optical parametric oscillator (see Fig. 1). Its output was focused on a cell containing sodium vapor. The pulse of radiation had a duration of ~ 30 nsec and a peak power of ~ 50–200 kW at a spectral width $\Delta\nu$ of 3 to 5 cm^{-1}. The sensitivity of the detection system at the frequency of the fifth harmonic reached $\sim 10^{-15}$J. To decrease the scatter of the

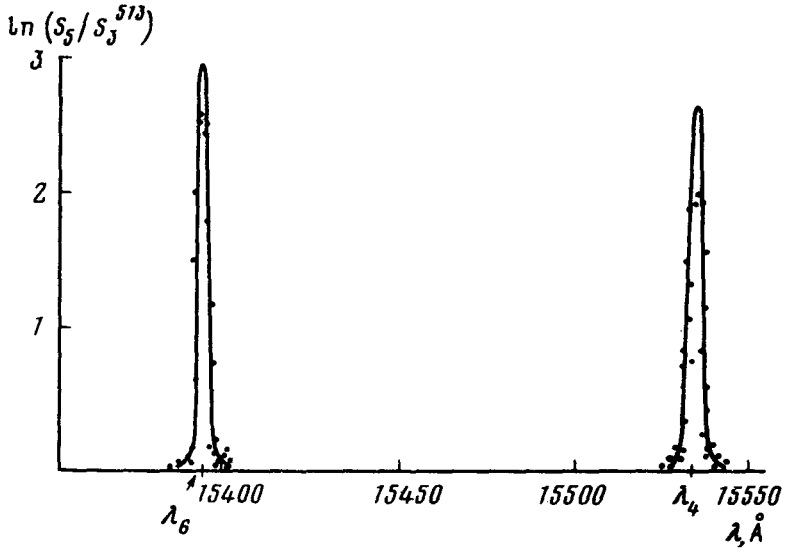

FIG. 2. Energy of fifth harmonic as a function of the wavelength of the radiation from the parametric oscillator.

experimental points due to uncontrollable fluctuations of the transverse intensity distribution in the pump beam, the fifth-harmonic signal S_5 was normalized to the third-harmonic signal S_3 generated in a $CaCO_3$ crystal in the reference channel.

4. We have experimentally investigated the dependence of the energy of the fifth harmonic on the wavelength λ of the emission of the parametric oscillator. When λ was tuned over the range 15550–15350 Å, a sharp increase in the conversion efficiency was observed near the four-photon resonance with the $3s$–$4s$ transition in atomic sodium ($\lambda_4 = 15535$ Å) and near the six-photon resonance with the $3s$–$8s$ transition ($\lambda_6 = 15393$ Å), as is shown in Fig. 2. Far from these resonances the signal of the fifth harmonic was below the sensitivity level of the detector. The conversion coefficient at the exact four-photon resonance was $\sim 10^{-10}$ at a pump power P_1 of about 200 kW.

5. The results obtained indicate that in our experiments the generation of the fifth harmonic is due to a six-photon process in the fifth-order nonlinear coefficient $\chi^{(5)}(5\omega)$ and an eight-photon combination $6\omega - \omega \rightarrow 5\omega$ in the nonlinear coefficient $\chi^{(7)}(6\omega - \omega)$ (see Fig. 3). We point out that the eight-photon process is the lowest-order process in which the observed six-photon resonance $3s$–$8s$ can occur.

In the calculations, the radiation of the parametric oscillator was modeled by Gaussian noise. We obtained the following formula for the power P_5 of the harmonic generated in the direct six-photon process:

$$P_5 \approx 9.6\,(10\lambda)^{-6} N^2 b^{-2} P_1^5 F\left(b\Delta k, \frac{b}{L}\right) |Q_m r_m|^2 I(\delta). \tag{1}$$

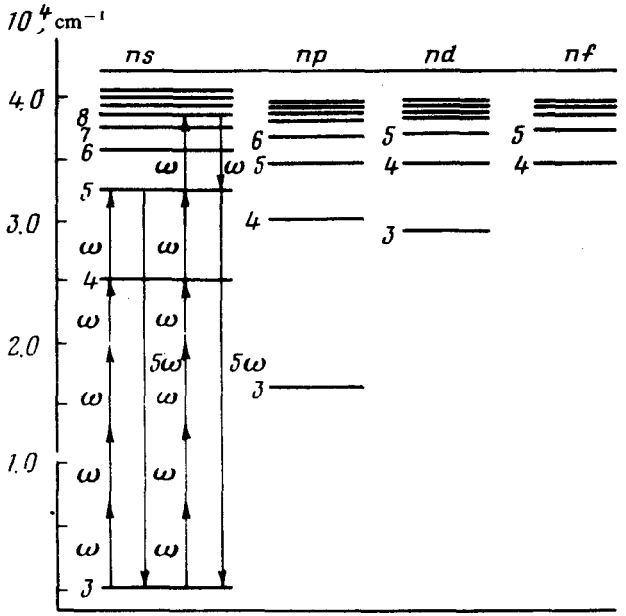

FIG. 3. Diagram of sodium levels and scheme of observed processes.

Here N is the concentration of sodium atoms, b is the confocal parameter, $\Delta k = k_5 - 5k_1$ is the wave-vector mismatch, L is the length of the nonlinear medium, F is the focusing function (see Ref. 7), and Q_m and r_m are matrix elements of the many-photon transitions between the levels of the m-photon resonance.[6] The function $I(\delta)$ depends on the frequency mismatch of the resonance $\delta = m\omega - \omega_{21}$. For $\delta \lesssim \Delta\omega = 2\pi c\Delta\nu$ it is the form

$$I(\delta) \approx m!(5-m)! \sqrt{\frac{\pi \ln 2}{m}} \frac{4}{\Delta\omega} \int_0^\infty \exp\left(-\frac{\gamma_0^2}{2} r^2 - 2\gamma r - \frac{\beta^2 \tau_0^2}{2\sqrt{m}}\right) \times \left[1 + \Phi\left(\frac{\sqrt{2}r}{r_0} - \frac{\beta^* \tau_0}{2\sqrt{m}}\right)\right] dr, \quad (2)$$

where γ_0 is the Doppler half-width of the line, γ is the homogeneous width of the transition, $\beta = \gamma + i\delta$, and $\tau_0 = 4(\sqrt{\ln 2})/\Delta\omega$. The factor $m!(5-m)!$ comes about because of the Gaussian statistics of the radiation. Analogous formulas hold for the eight-photon process.

For the four-photon resonance we obtained good quantitative agreement between the experimental and theoretical data. In the calculations we considered that for our experiment $N \simeq 0.8 \times 10^{17}$ cm^{-3}, $b \simeq 4$ cm, $L \simeq 8$ cm, $F(b\Delta k, b/L) \simeq 0.1$ (Ref. 7), and $m = 4$. The matrix elements Q_4 and r_4 were evaluated with the dipole moments given in Ref. 8. The calculated value $|Q_4 r_4| \simeq 3.6 \times 10^{-30}$ cgs esu is in good agreement with the experimental value $|Q_4 r_4| \simeq 2.5 \times 10^{-30}$ cgs esu.

For the eight-photon process there are no data on the focusing function, so the comparison of the theory with experiment is of qualitative nature: the calculated curve 2 in Fig. 2 "correlates" with the experimental value at the exact six-photon resonance. It is seen that the theoretical curve is in good agreement with experiment.

There can also be a contribution to the fifth-harmonic signal from cascade processes having resonant gain in the effective nonlinearity $3\omega \to 3\omega + \omega + \omega \to 5\omega$ for the four-photon resonance, and correspondingly $3\omega \to 3\omega + 3\omega - \omega \to 5\omega$, $3\omega 3\omega + \omega + \omega + \omega - \omega \to 5\omega$, and $5\omega \to 5\omega + \omega - \omega \to 5\omega$ for the six-photon resonance. However, it is in principle unimportant for spectroscopic probing of many-photon resonances what sort of process, direct or cascade, produces the observed signal.

In conclusion, we remark that higher-order frequency-combination processes enable one to study quantum transitions which are unaccessible by the methods of two-photon spectroscopy and four-wave mixing spectroscopy. For example, six-photon parametric processes by means of which one can probe from the ground state of the f level in alkali-metal atoms. The use of pump radiation at several frequencies will no doubt extend the spectroscopic capabilities of these methods. The use of picosecond pulses will permit observation of transient many-photon resonance effects, such as the damping of free polarization, many-photon nutation, etc. We have already obtained preliminary results in this area.[1)]

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[1)]We have done experiments with picosecond pulses in cooperation with co-workers at Vilnius State University. The results are to be published.

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