Two-photon excitation of cesium atoms by correlated optical fields

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The efficiency of two-photon excitation of cesium atoms in the $6D_{3/2,5/2}$ state by nonmonochromatic frequency- and phase-correlated optical radiation was investigated experimentally. The correlation between the harmonics of the noise jump field was produced by nonlinear optical mixing of the initial wide-band radiation and the narrow-band radiation from a laser heterodyne. It was found that the effect of the correlated fields on resonance systems (cesium atoms) was independent of the width of the noise spectrum, which was varied by a factor of more than 100.

1. It is well-known that the correlation of nonmonochromatic radiation plays an important part in nonlinear processes in which such radiation interacts with elementary systems. There have been a number of studies in which the effect of the correlation properties of radiation on the efficiency of multiphoton excitation of resonance systems was investigated. The effect of correlation between adjacent channels (cross-correlation) on the two-step excitation of a three-level system by radiation with diffusing-phase statistics was investigated in Ref. 1. Cross-correlation is suggested in Ref. 2 as a means of reducing noise in systems using Raman scattering of laser radiation, when the noise is due to fluctuations in the laser frequency. An implementation of this idea is reported in Ref. 3. The effect of correlation properties of fields with a diffusing phase on two-photon absorption of radiation was observed in Ref. 4. The width of the two-photon absorption spectrum was found to depend on its correlation properties for a noise spectrum of fixed width.

Bonch-Bruevich *et al.*[5] have shown that the efficiency of interaction with wide-band radiation can be raised to the corresponding efficiency for monochromatic radiation in the case of two-photon excitation of a two-level system by establishing a particular correlation between the harmonics of the nonmonochromatic noise field. Correlation was attained by nonlinear mixing of nonmonochromatic radiation from a heterodyne (ω_h) with an initial noise radiation whose frequency lay in the neighborhood of a frequency ω_0. A second noise field was generated in this way and its harmonics, lying in the neighborhood of ω_i, were strictly related to the harmonics of the initial noise field. We used a radiofrequency field with wavelength much greater than the linear dimensions of the region filled with the ensemble of two-level systems (we investigated transitions between the Zeeman sublevels of the ground state of Cd atoms). The instantaneous fields can therefore be regarded as identical for all the atoms, and questions relating to the propagation of the radiation can be ignored. It is clear that these conditions are not satisfied in the optical range for which the interaction between resonance systems and wide-band fields with correlated harmonics requires separate analysis. We have demonstrated experimentally that the effect of wide-band radiation on resonance systems can be amplified in the optical range by correlating its harmonics.

2. We have investigated the following two-photon transitions in Cs atoms:

$$6S_{1/2}\to 6P_{1/2,\,3/2}\to 6D_{3/2},$$
$$6S_{1/2}\to 6P_{3/2}\to 6D_{5/2}.$$

Figure 1 illustrates the energy-level diagram and the optical transitions on cesium atoms. It defines the spectral range of the pump radiation, the positions of the intermediate levels, and the oscillator strengths of the transitions. The transitions were chosen because of their large oscillator strengths, the possible overlap of the corresponding spectral range with laser radiation having the necessary parameters, and the practical feasibility of generating fields with correlated harmonics in the optical range, using a scheme analogous to that employed in Ref. 5 and based on the nonlinear mixing of two waves in a crystal.

Figure 2 shows a block diagram of the apparatus employed. The initial wide-band (noise) radiation was produced by a single-mode dye laser (mean wavelength $\lambda_0 = 930$ nm). The dye was pumped by pulses from the ruby laser 1 at a repetition rate of 1/7 Hz with 0.8–1 J/pulse. When 100–1000 statistically independent modes were generated, the result was a radiation field with statistics approaching complex Gaussian noise.[6] The aim of the experiment was to investigate the efficiency of two-photon excitation as a function of the width of the noise spectrum in the presence of frequency and phase correlation. The optimum solution is to use laser-generated noise radiation with adjustable spectral

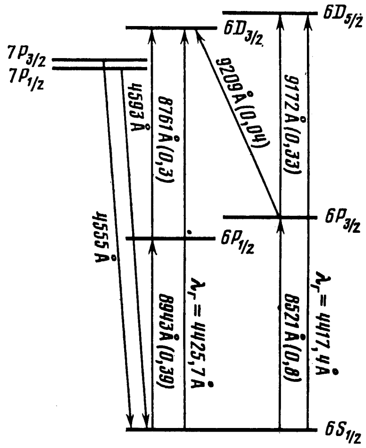

FIG. 1. Energy-level scheme of the cesium atom.

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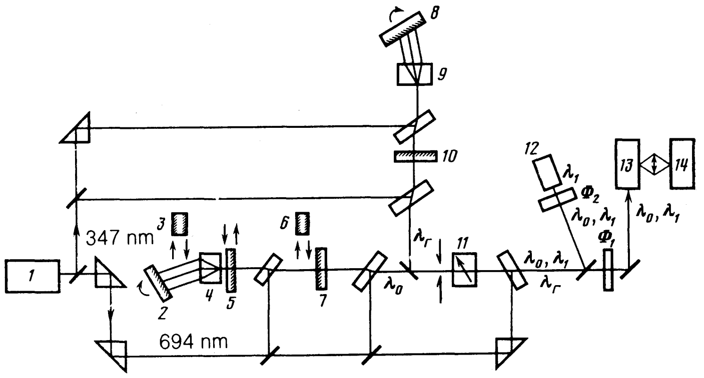

FIG. 2. Block diagram of the apparatus: 1—source of radiation with $\lambda = 694$ and 347 nm; 2, 8—diffraction grating; 3, 6—Fabry-Perot interferometers; 4,9—prism beam expanders; 5—mirror with $R = 100\%$, 7, 10—exit mirrors; 11—$LiIO_3$ crystal mixer; 12, 14—photodetectors; 13—cell containing cesium vapor.

width and constant ray propagation geometry. This was achieved by employing coaxially adjusted cavity resonators. The first resonator consisted of the diffraction grating 2 and mirror 7, whereas the second consisted of mirror 5 (reflectivity $R = 100\%$) and mirror 7. The two resonators were thus inserted one into the other. Mirror 5 can be displaced parallel to itself, or removed from the resonator without loss of adjustment. The width of the spectrum generated in the first resonator was 3 cm^{-1}, whereas the width in the second resonator was 80 cm^{-1}. Whenever necessary, the linewidth could be compressed down to 0.5 cm^{-1} by inserting the Fabry-Perot interferometer 3 with $t = 1$ mm into the first resonator. By inserting the interferometer 6 with $t = 20\,\mu m$ or $4\,\mu m$ into the second resonator, it was possible to produce intermediate linewidths of 20 cm^{-1} or 30 cm^{-1}, respectively. When the width of the spectrum was varied from 0.5 cm^{-1} to 80 cm^{-1}, it was found that the energy per pulse changed by a factor of about 3 and, on average, amounted to 0.05–0.1 J ($I_0 = 2\times10^{-8}$–3×10^{8} W/cm^2.

To produce the stochastic field with correlated harmonics, the wide-band radiation generated by the dye laser was directed onto the $LiIO_3$ crystal 11, in which it was nonlinearly mixed with the narrow-band radiation from the laser heterodyne of frequency ω_h. This produced the wide-band radiation in which the frequency ω_i and phase φ_i of each harmonic was related to the frequency ω_0 and phase φ_0 of the corresponding harmonic in the initial noise field by $\omega_i + \omega_0 = \omega_h, \varphi_i + \varphi_0 = \varphi_h$. Together with the initial field (radiation transmitted by the crystal without transformation), this results in a stochastic field with frequency and phase-correlated harmonics.

The laser heterodyne was a dye laser (Kumarin-120), pumped by the second harmonic of the ruby laser 1. The heterodyne wavelength $\lambda_h = 442$ nm and spectral width $\Delta\nu_h = 0.5$ cm^{-1} were produced in a resonator consisting of the diffraction grating 8 and exit mirror 10 ($R = 50\%$). The output energy of the laser-heterodyne pulse was 3×10^{-3}–5×10^{-3} J. This scheme ensured collinear propagation of radiation at frequencies ω_0, ω_1, and ω_h. A light filter Φ_1 (KS-19 glass) was placed in front of the cell containing the cesium vapor and completely removed the heterodyne radiation. Special tests were made to verify that the spectral widths of the transformed and original wide-band radiation were the same. Particular attention was paid to ensure maximum intensity I_1 of transformed radiation, so as to achieve the optimum ratio of this intensity to the initial optimum intensity I_0 for two-photon excitation. The optimum situation obtains when $I_0 f_{35} = I_1 f_{13}$, where f_{35} and f_{13} are the oscillator strengths of the first and second transition, respectively (see Fig. 4b below). Since, after the nonlinear crystal, the intensity of the transformed radiation I_1 for completely overlapping rays was only 4% of I_0, the transformed radiation was transmitted by the dye laser (maximum gain at 840 nm; pumped by the ruby laser radiation). The final result was $I_0 f_{35}/I_1 f_{13} = 1{:}2.5$. The optimum conditions for two-photon excitation were therefore not satisfied, but experiments showed that the arrangements were good enough to produce reliable results.

In accordance with the level scheme for cesium atoms shown in Fig. 3, excitation to the $6D_{3/2,5/2}$ state should result in emission due to transitions from these levels to the intermediate states $6P_{1/2,3/2}$. The emission lies in the infrared and its wavelengths ($\lambda = 8761$ Å, 9208 Å, 9172 Å) are close to the wavelength of the pump radiation in the upper channel. This gives rise to difficulties in recording the two-photon excitation because the signal due to transitions from the $6D_{3/2,5/2}$ states is difficult to filter off from the scattered pump radiation.

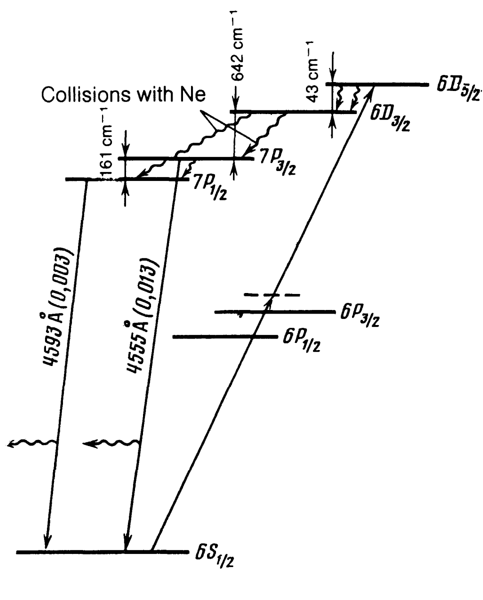

FIG. 3. Transfer of excitation in cesium during collisions with buffer-gas atoms (Ne).

This difficulty can be obviated by adding a buffer gas (Ne, Xe, etc.) to the cesium vapor. Collisions between cesium atoms that have been subjected to two-photon excitation and the buffer gas result in de-excitation from the $7P_{1/2,3/2}$ states, producing radiation in the visible range (4593 Å, 4555 Å). Observations have shown that energy transfer from the one state ($6D$) to the other ($7P$) is very effective in inelastic collisions. For example, when the lifetime of the $6D$ excited state is 3×10^{-8} s, and the buffer-gas pressure is 100 Torr (Ne or Xe), the large inelastic cross section ($\sigma(6D\rightarrow7P)=2.5\times10^{-16}$ cm^2) ensures that the probability of this process is close to unity.

The cesium lines that appear in the blue part of the spectrum as a result of the de-excitation are quite far from the infrared pump lines, and can be almost completely filtered off by absorbing light filters transparent in the blue-green range. They fall into the region of high spectral sensitivity of the photomultiplier used to record the radiation (14 in Fig. 2). This method could be readily used to record the two-photon excitation signal in the absence of background illumination.

3. Before the correlation effects in two-photon excitation of cesium atoms by optical noise fields were investigated, we carried out control measurements under monochromatic (narrow-band) excitation of the atomic systems, and selected optimum parameters and working conditions for the components of the apparatus. The observed signal corresponding to the two-photon excitation of cesium atoms in the $6D_{3/2}$ and $6D_{5/2}$ states in a narrow-band (3 cm^{-1}) single-frequency field (Fig. 4a) was compared with theoretical estimates of the two-photon excitation probability. Next, we used the two-frequency narrow-band fields (Fig. 4b) produced by the heterodyne system to measure the signal due to the strongest resonance, which was then compared with the signal produced in the single-frequency field. The temperature of the cell containing the cesium vapor was controlled by a regulating device. The temperature of the coldest part of the cell (finger) was 130 °C ($P_{Cs}=3\times10^{-3}$ Torr), and the temperature variations did not exceed $\pm$ 4 °C.

We now present a brief summary of our measurements. The theoretical estimate of the ratio of excitation probabilities W_{ik} for the transitions shown in Fig. 4a is

$$\frac{W_{14}(6D_{3/2},\lambda=8851\ \text{Å})}{W_{15}(6D_{5/2},\lambda=8835\ \text{Å})}=\frac{f_{12}f_{24}}{f_{13}f_{35}}\left(\frac{\Delta_2}{\Delta_1}\right)^2\approx\frac{0.12}{0.25}\left(\frac{400}{118}\right)^2\approx5.3$$

where f_{ik} is the oscillator strength and Δ_1 and Δ_2 (cm^{-1}) are the resonance defects corresponding to the intermediate levels. The measured ratio was 4–5, which agrees with the theoretical prediction to within $\pm$ 25%.

Without changing the pump-ray propagation geometry, we then replaced the dye in the main laser, shifted the wavelength λ_0 with the aid of the diffraction grating to the 930 nm region, and made the heterodyne emit at 442 nm. A difference wave was produced (840 nm), and two-photon excitation was examined for a narrow-band pump at two wavelengths. By retuning λ_h within 8 Å (41 cm^{-1}), we were able to transfer from one two-photon resonance to the other.

The theoretical estimate of the efficiency ratio for the two-photon excitation in the case of the transitions indicated in Fig. 4b is

$$\frac{W_{15}(6D_{5/2})}{W_{14}(6D_{3/2})}=\frac{f_{13}f_{35}}{f_{13}f_{34}}\left(\frac{\delta_1}{\delta_2}\right)^2=\frac{0.33}{0.04}\left(\frac{120}{162}\right)^2\approx4.5.$$

Experiment yielded a figure that was higher than this by a factor of two. It may be that this discrepancy is due to a considerable fraction of circularly polarized radiation in the pump, which was ignored in the above theoretical estimate.

The cesium atoms were then pumped by the uncorrelated noise radiation to the $6D_{3/2}$ state (Fig. 4a) and by the correlated radiation to the $6D_{5/2}$ state (Fig. 4b). For both two-photon excitation schemes indicated in Figs. 4a and b, we compared the efficiencies and the agreement or otherwise with theoretical estimates:

$$\frac{W_{15}(\lambda_0,\lambda_1,6D_{5/2})}{W_{14}(\lambda,6D_{3/2})}=\frac{I_0I_1}{I^2}\frac{f_{13}f_{35}}{f_{12}f_{24}}\left(\frac{\Delta_1}{\delta_2}\right)^2.$$

where I is the intensity under single-frequency interaction and I_1, I_0 are the intensities under two-frequency interaction.

These measurements show that the theoretical estimates agree to within 30% with the experimental data on the radiation emitted under two-photon excitation.

4. Our main result is the comparison between the efficiency W_{15} of two-photon excitation of cesium in the fre-

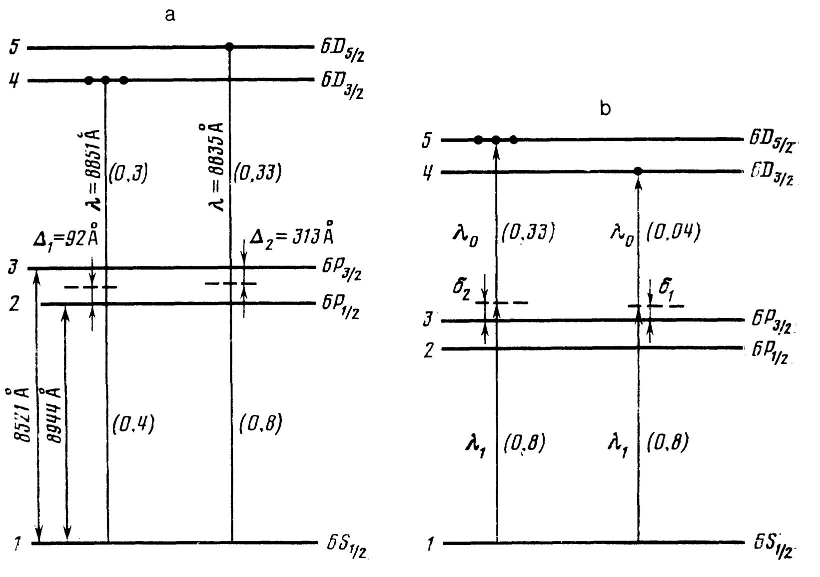

FIG. 4. Two-photon single-frequency (a) and two-frequency (b) excitation.

quency and phase correlated optical noise fields and the efficiency W_{14} for uncorrelated noise fields. In our experiments, we used correlated and uncorrelated fields with spectral widths $\Delta_n = 0.5$, 3, 20, 30, and 80 cm^{-1}. We recorded the emitted intensity S under two-photon excitation, the intensity I_1 of the difference wave, and the resonance defect δ for each width of the noise spectrum. In the subsequent comparison of the emission signals for different spectral widths Δ_n, all the signals were normalized to the square of I_1 and the square of δ:

$$Z(\Delta_n) = S/I_1^2\delta^2.$$

Experimental results obtained for correlated and uncorrelated fields, averaged over a number of measurements, are compared below with theoretical calculations for uncorrelated fields:

correlated fields—Z (0.5 cm^{-1}):Z (3 cm^{-1}):Z (20 cm^{-1}):Z (30 cm^{-1}):Z (80 cm^{-1}) = 1:1:1:0.93:0.17;

uncorrelated fields (theory)—Z (0.5 cm^{-1}):Z (3 cm^{-1}):Z (20 cm^{-}):Z (30 cm^{-1}):Z (80 cm^{-1}) = 1:0.17:0.025:0.017:0.007;

uncorrelated fields (experiment)—Z (0.5 cm^{-1}):Z (3 cm^{-1}):Z (80 cm^{-1}) = 1:0.17:0.005.

For stochastic wide-band fields, the efficiency W_{14} of two-photon excitation of resonance systems is inversely proportional to the spectral width of the radiation, in complete agreement with theory.

On the contrary, for correlated wide-band fields, the two-photon excitation signal is practically independent of the spectral width Δ_n. This agrees with previous experimental data, obtained in the radiofrequency range.[5] There is a noticeable departure of the magnitude of Z in correlated fields from the practically constant value for $\Delta_n = 80$ cm^{-1}. The reasons for this are still unclear and will require further investigation.

Our results confirm and experimentally demonstrate that resonance systems can be made to radiate in the optical band under wide-band excitation by correlated fields.

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