Resonance fluorescence spectrum of a dense three-level medium (sodium vapor) in an intense laser field

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Spectra of the resonance fluorescence of a dense three-level medium in an intense laser field have been measured for the first time. The density of the medium was $N \sim 10^{15}$ cm^{-3}. The spectra are found to have a multicomponent structure, which results from a Rabi splitting into three quasilevels of both the $3S$ ground level and the two $3P$ excited levels of the sodium atom. Spectral calculations carried out here agree well with experimental data when allowance is made for spatial variations of the laser beam and for the absorption of fluorescence photons in the unperturbed sodium vapor.

Research on the spectra of the fluorescence which arises as a result of the application of a resonant laser beam is playing a fundamental role in efforts to reach an understanding of processes by which radiation interacts with matter. For a low-density, two-level medium, this spectrum consists of a symmetric triplet whose side components are separated from the central peak by a distance equal to the generalized Rabi frequency.[1-4] In dense media, in which collision processes are important, the center of gravity of this multiplet shifts toward the frequency of the resonance transition.[5,6] At high intensities of the laser beam, however, the two-level description breaks down for real atomic systems (e.g., the vapor of an alkali metal), since the distance to the perturbing neighboring levels becomes comparable to the Rabi frequency. In this letter we report the first experimental results on the observation of resonance fluorescence in an intense laser field in which a multicomponent structure has been observed in the emission spectra of the resonant medium. This structure arises from a Rabi splitting into three quasilevels of both the $3S$ ground level and the two excited $3P$ levels of the sodium atom (Fig. 1). The magnitude of the splitting is determined in the approximation of a three-level medium by a characteristic equation which is a consequence of the solution of a system of time-dependent Schrödinger equations in the resonance approximation:[7]

$$\epsilon(\epsilon+\Delta_2)(\epsilon+\Delta_3)-1/4(\mu_{12}E/\hbar)^2(\epsilon+\Delta_3)-1/4(\mu_{13}E/\hbar)^2(\epsilon+\Delta_2)=0. \qquad (1)$$

Here $\Delta_2=\omega_{12}-\omega_L$; $\Delta_3=\omega_{13}-\omega_L$; ω_L is the frequency of the laser light; μ_{12}, μ_{13}, ω_{12}, and ω_{13} are respectively the dipole moments and unperturbed resonant frequencies of the $D_1(3^2S_{1/2}-3^2P_{1/2})$ and $D_2(3^2S_{1/2}-3^2P_{3/2})$ transitions in the sodium atom; E is the electromagnetic field of the laser wave; and $\hbar$ is Planck's constant. The roots of this equation, ϵ_k ($k=1,2,3$), are Rabi oscillation frequencies (quasienergies). They determine generalized Rabi frequencies for the three-level system:

$$\Omega'_j=|\epsilon_m-\epsilon_n| \quad (j=1,2,3; \quad m\neq n). \qquad (2)$$

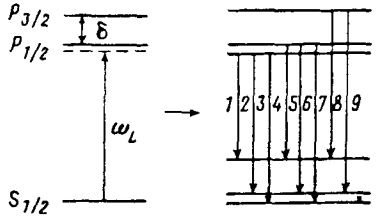

FIG. 1. Shift and splitting of atomic energy levels in an intense laser wave. 3,5,7—Transitions at the laser frequency, ω_L; 6,2—transitions at the frequencies $\omega_1^{\pm}=\omega_L\pm\Omega_1'$; 8,4—at the frequencies $\omega_2^{\pm}=\omega_L\pm\Omega_2'$; 9,1—at the frequencies $\omega_3^{\pm}=\omega_L\pm\Omega_3'$.

It follows from Fig. 1 that the fluorescence spectrum of the three-level medium should contain up to six components, with frequencies satisfying $\omega_j^{\pm}=\omega_L\pm\Omega_j'$, along with three degenerate components, with frequencies $\omega_0=\omega_L$. In the weak-field limit (the limit of a small Rabi frequency, $\Omega_i=\mu_{1i}E/\hbar\ll\Delta_i$, $i=2,3$), the generalized Rabi frequencies are $\Omega_1'\simeq\delta$ ($\delta=\omega_{13}-\omega_{12}$), $\Omega_2'\simeq\Delta_2$, and $\Omega_3'\simeq\Delta_3$. The corresponding transitions describe the spectrum of various well-known types of Raman scattering (Refs. 8 and 9, for example).

The experimental apparatus consisted of the test cell holding sodium vapor, a dye laser, and a variety of diagnostic instruments. The heated cell, similar to that described in Ref. 10, made it possible to produce a cylindrical column of sodium vapor with a density of 10^{13}–10^{16} cm^{-3} and a height of 1.5 cm. The number density of sodium atoms, N, and its radial profile in the cell, $N(r)$, were found through an Abel transformation of measurements of $\int N(r)dr$ with a Michelson interferometer by the Rozhdestvenskiĭ-hook method. The interferometer was illuminated by a wide-band auxiliary dye laser. Before the experiment, the cell was evacuated to a pressure of 10^{-5} torr. An inert gas (argon) was then admitted to a pressure $\sim$ 0.3–5 torr, and the cell was heated to the desired temperature. Measurements showed that the diameter of the vapor column at the half-maximum points of the distribution was 8–10 cm, and that it depended only weakly on the temperature and pressure of the buffer gas.

The tunable dye laser (the dye was rhodamine 6G) was pumped by the second harmonic of a Nd:YAG laser operating at a frequency of 10 Hz. The dye laser generated linearly polarized light with an energy E_0 up to 6 mJ at a spectral width of 0.008 nm and a pulse length $\tau=18$ ns. The laser beam was focused by a lens with a focal length of 45 cm at the center of a cell, intersecting the column of sodium vapor along a diameter of this column. The energy profile $E(r)$ of the laser beam along the beam radius r was measured by a linear array of photodiodes in the focusing region. It was found that this profile can be approximated well by $E(r)\propto[r_0^2/(r_0^2+r^2)]^{1.5}$, where $r_0=0.4$ mm. The beam radius at half-maximum of the energy distribution was $r_{1/2}=0.3$ mm in this region. The intensity of the laser light in the experiments was calculated from the formula $\mathcal{F}=E_0/(\tau S)$, where $S=\pi r_{1/2}^2$.

The fluorescence of the sodium vapor which is induced by the laser light is collected from the center of the cell in a direction perpendicular to the axis of the laser beam by two objective lenses. These lenses form an image of the emitting region on the entrance slit of a diffraction monochromator (1200 lines/mm). An FÉU-84 photomultiplier is positioned behind the exit slit of this monochromator. The output signal from this photomultiplier is detected by a digital sampling voltmeter at the time

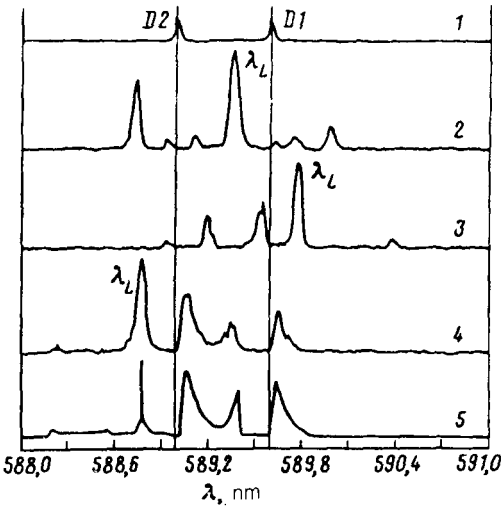

FIG. 2. Experimental (*2–4*) and theoretical (*5*) fluorescence spectra of sodium vapor ($N=2.3\times10^{15}$ cm^{-3}, $\mathcal{F}=20$ MW/cm^2). *1*—Calibration spectrum of a sodium lamp; *2*—$\Delta\lambda_L=-0.37$ nm; *3*—$\Delta\lambda_L=-0.79$ nm; *4*, *5*—$\Delta\lambda_L=0.21$ nm, $\Delta\lambda_L=\lambda_{D2}-\lambda_L$.

corresponding to the crest of the output pulse from the dye laser. Within the strobe pulse (4 ns), the generation intensity was essentially constant. To record the fluorescence spectra, the transmission wavelength of the monochromator was automatically scanned in the vicinity of the sodium D_1 and D_2 lines. The spectral resolution of this measurement system was 0.02 nm.

Figure 2 shows the fluorescence spectra of sodium vapor measured in these experiments. It can be seen from these results that if the laser wavelength λ_L lies between the D lines, then the fluorescence spectrum has seven components, reflecting the splitting (described above) of the three-level medium in the intense laser field. If λ_L instead lies outside this interval, then the spectrum contains only five components. A characteristic feature of these spectra is that when the laser light lies on the high-frequency side of the D_2 line, the most intense fluorescence lines are at frequencies lower than the laser frequency, while in the case of a low-frequency detuning from the D_1 line we find the opposite behavior. The intensities of the two other components are extremely low in this case (as was confirmed by calculations, as discussed below), at the noise level. The observed asymmetry of the fluorescence spectra at sufficiently high pressures of the buffer gas and the sodium vapor was due to the important role played by collisions, as was mentioned above.[5,6] Note, however, that in the three-level medium, in contrast with the two-level case, the fluorescence spectra do not have a strictly symmetric shape even in the absence of collisions.[11]

The detuning $\Delta\lambda_j=\lambda_L-\lambda_j$ of the wavelength of the peak of the fluorescence components detected, λ_j, is shown as a function of $\Delta\lambda_L=\lambda_{D2}-\lambda_L$ (λ_{D2} is the wavelength of the sodium D_2 line) in Fig. 3. Also shown here are the positions of the detunings of the components calculated from Eqs. (1) and (2) for an intensity of 20 MW/cm^2 (the dashed curve), which corresponds to the experimental conditions. It follows from these results that, over the entire detuning range, the measured positions of the components are shifted a significant distance away from the theoretical positions

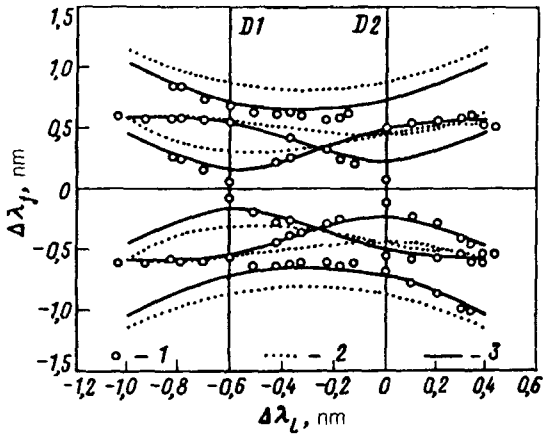

FIG. 3. Experimental (points) and theoretical (solid and dotted curves) results on the shift of the components of the fluorescence spectrum of sodium vapor. $N=2.3\times10^{15}$ cm^{-3}. Points and dotted curves—$\mathcal{F}=20$ MW/cm^2; solid curves—5 MW/cm^2.

and correspond roughly to a much lower intensity, $\mathcal{F}=5$ MW/cm^2. A corresponding theoretical curve is also shown in Fig. 3 (the solid curve). There are several factors which might explain this discrepancy. On the one hand, the shifts of the fluorescence components (and also the spectral widths of these components) observed experimentally are affected by spatial variations of the laser beam along the observation path, so the generalized Rabi frequency Ω_j' is also spatially nonuniform. Because of the saturation of the fluorescence at large values of $\mathcal{F}$, the spatial wings of the laser beam, in which the intensity is relatively low, affect appreciably the shape of the measured spectra. On the other hand, the shape of the latter spectra is distorted by the absorption of fluorescence photons in the dense sodium vapor unperturbed by the laser light. Simple estimates show that this absorption can be fairly strong, even though the frequencies of the components are detuned fairly far from the resonance components. In addition, the phase self-modulation of the laser light is important. It is manifested particularly clearly in a broadening of the unshifted fluorescence component at the frequency of the laser light. However, time-resolved experiments allowed us to avoid a change in the generalized Rabi frequency Ω_j' during the laser pulse and to avoid a shift of the fluorescence lines toward the unperturbed transition frequencies in the afterglow of the vapor.

In an effort to explain these results, we have carried out some steady-state numerical calculations of the fluorescence spectra of sodium vapor in the three-level approximation and also under the assumption of a δ-function spectrum of the exciting laser light. We used the theoretical model developed in Ref. 11, which is based on the formalism of an atomic-photon density matrix.[12-14] In modeling the spectra we used the constants given in Refs. 15 and 16 for the broadening induced by the buffer gas and for the resonant broadening. The calculations also incorporated the strongest effects which lead to a deformation of the observed spectra, the spatial nonuniformity of the laser beam, and the absorption of fluorescence photons in the unperturbed vapor. Curve 5 in Fig. 2 shows the results of these calculations for the case $\Delta\lambda_L=0.21$ nm. These results agree well with the experimental spectrum, except with regard to the unshifted fluorescence line at the frequency of the exciting light. We know that there

are two components in the intensity of this line: a coherent one and an incoherent one (Refs. 1 and 5, for example). Since the calculations ignored the multifrequency nature of the exciting light and also the finite resolution of the monochromator, the calculated width of the incoherent component turned out to be on the order of the collisional width and cannot be seen at the scale of this figure. The amplitude of this component is considerably greater than the amplitudes of the lines of the other components. The spectral intensity of the coherent component, on the other hand, is in general proportional to $\delta(\omega-\omega_L)$. For both reasons, the amplitude of the unshifted fluorescence line is shown only schematically in the theoretical spectrum.

In summary, this study has yielded the first fluorescence spectra of a dense three-level medium in an intense laser field. It has been found that these spectra have a multicomponent structure, which stems from a Rabi splitting into three quasilevels of the ground level and the two $3P$ excited levels of the sodium atom. The asymmetry of the fluorescence spectra found experimentally is determined by collisions and also by the fundamentally multilevel nature of the atomic system studied here. The calculations carried out as part of the present study agree well with the experimental data. Nevertheless, the complete validity of the steady-state approach in calculations describing fluorescence under the conditions of these experiments remains an open question.

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