Optical pumping and nonlinear effects in the spectroscopy of the cesium D_2 line

Yu. A. Bykovskiĭ, V. L. Velichanskiĭ, V. K. Egorov, A. S. Zibrov, and V. A. Maslov

Moscow Engineering Physics Institute
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We report here the first use of a semiconductor laser for nonlinear spectroscopy within Doppler-broadened lines. Dips were observed in the dependence of the photoluminescence intensity $\Phi(\nu)$ of Cs^{133} vapor on the generation frequency (ν) of the exciting laser.

The semiconductor-laser generation frequency was tuned by modulating the injection current.[1,2] The radiation of the laser (1, Fig. 1) was shaped into a beam with angular aperture $\phi = 4°$, passed through a cell (2) of length 1.8 cm filled with cesium vapor ($T = 293$ °K, atom concentration $N = 2.8 \times 10^{10}$ cm^{-3}), and was reflected by a mirror (3). The luminescence signal from the photomultiplier (4) was fed to an oscilloscope. The oscillograms of Fig. 2 show plots of $\Phi(\nu)$ with the laser frequency varied in the region of the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}$ absorption line in the absence (a) and in the presence (b) of the reflected wave, and in the latter case also at a higher laser intensity (c). The time of variation of the frequency within the limits of the line width (500 MHz) was 5 msec. Two dips are seen on the long wave wing of $\Phi(\nu)$.

In the absence of nonlinear effects, the contour of $\Phi(\nu)$ coincides with the contour of the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}(F=3,4,5)$ absorption line and is determined by the parameters given in Fig. 3. Let us examine the mechanisms capable of causing nonlinearity of the luminescence and the formation of the dips.

1. In a two-level system, nonlinearity arises as a result of redistribution of the level populations. The condition $I\sigma\tau < 10^{-2}$ (τ is the lifetime of the atom in the excited state, I is the proton flux density, and σ is the absorption cross section) was satisfied in the experiment. Consequently, the dip for which the given mechanism is the only one possible, at the frequency ν_3, does not appear.

2. Nonlinear effects can occur in alkali-metal vapors also as a result of the redistribution of the populations of the sublevels of the ground state through optical pumping (OP). In a system of atoms having the energy spectrum of Fig. 3d, the populations of the levels of the ground state at a given frequency of the laser light are altered by the OP only in a group of atoms having a definite velocity projection V_x on the light propagation direction. The distribution of the atoms over the velocities and the sublevels of the ground state in the presence of two opposing laser-radiation beams of equal intensity is shown in Fig. 3b (the laser frequency does not coincide with the line center). When the laser frequency passes through the line center, a dip is produced in the $\Phi(\nu)$ contour, with a depth $\sim (1/\tau)[2p(I) - p(2I)]$, where p is the population of the $6P_{3/2}$ level. The nonlinearity of $p(I)$ leads to the formation of the dip. The relaxation time of the populations n_3 and n_4 was determined in our case by the time of flight of the atoms through the laser beam ($T \approx 10^{-5}$ sec). Thus, the laser-radiation density at which a noticeable bleaching due to the OP appears ($I = 1/\sigma T \approx 10^{14}$ cm^{-2} sec^{-1}) is smaller by three orders of magnitude than the density necessary to bleach a two-level system.

3. If the excited state has two levels (Fig. 3e), the distance between which is smaller than $\Delta\nu_D$, then a "crossover" dip[3] appears at the frequency $(\nu_1 + \nu_2)/2$, and is due to OP in our case.

When the generation frequency is varied slowly, a stationary regime is established for atoms with definite V_x (this condition is satisfied in the experiment), and the limiting value of the luminescence power is determined by the influx of the unexcited atoms into the light beam, i.e., by a quantity on the order of n/T, where $n = (\Delta\nu_{nat}/\Delta\nu_D)2\sqrt{\ln 2} \cdot N$ is the concentration of the atoms that interact effectively with the monochromatic radiation. This means that in OP the luminescence intensity, and consequently also the depth of the dips in $\Phi(\nu)$, are limited to a value proportional to n/T.

4. Let us examine the level system of Fig. 3f (one of the optical transitions is forbidden). In this case, a

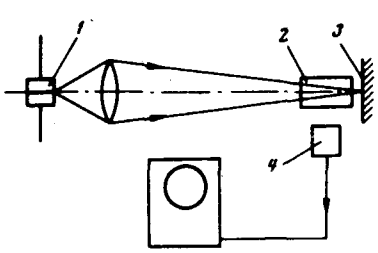

FIG. 1. Diagram of setup.

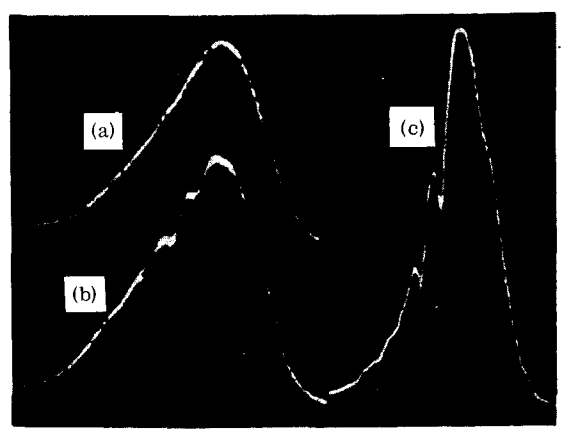

FIG. 2. Oscillograms of the luminescence of Cs^{133} vapor as the semiconductor laser frequency is varied in the region of the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}$ line.

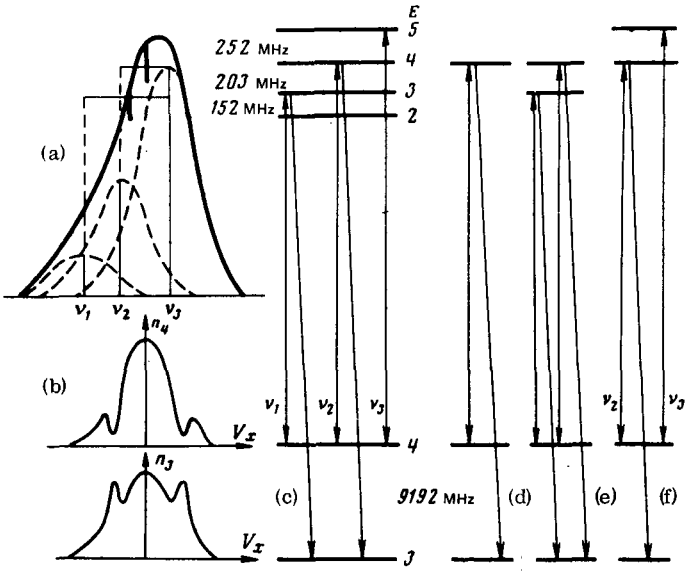

FIG. 3.

crossover dip is observed in the strong luminescence line (ν_3), and its relative depth can reach values close to unity. Indeed, if $|\nu-(\nu_2-\nu_3)/2| > \Delta\nu_{nat}$, then four groups of atoms interact with the radiation. Two of them, in which there is no OP $[V_x=\pm(\nu-\nu_3)c/\nu]$ produce strong spontaneous radiation proportional to $I\sigma n$. In the two others $[V_x=\pm(\nu-\nu_2)c/\nu]$, the upper sublevels of the ground state are depleted, and their contribution to the luminescence is small ($\sim n/T$). The light of frequency $\nu=(\nu_2+\nu_3)/2$ interacts with only two groups of atoms $[V_x=\pm(\nu_3-\nu_2)c/(\nu_3+\nu_2)]$. In each of these groups, the atoms are transferred by the OP to a non-absorbing state, and this is the cause of the deep dips in the $\Phi(\nu)$ dependence.

As seen from the level scheme of the cesium atoms, two crossover dips are possible in the $\Phi(\nu)$ dependence at the frequencies $(\nu_1+\nu_3)/2$ and $(\nu_2+\nu_3)/2$, and occur with simultaneous participation of transitions with and without OP. These are precisely the dips observed in the experiment, as confirmed by their positions on the $\Phi(\nu)$ line. The ratios of the frequency interval between the dips to the line width are 0.21 (Fig. 2b) and 0.25 (Fig. 2c). The distance between the vertices of the dips, $(\nu_2+\nu_3)/2-(\nu_1+\nu_2)/2=(\nu_2-\nu_1)/2\approx 100$ MHz, and was used to determine the frequency scale. The widths of the $\Phi(\nu)$ lines in Figs. 2b and 2c are 480 and 400 MHz, respectively. The line-width decrease accompanying the increase in the dip depth is due to the bleaching of the cesium vapor by the OP at the frequencies of the long-wave components.

The difference between the observed width of the dips (45 ± 5 MHz) and the natural width (5 MHz) is due to nonparallelism of the beam ($\phi\Delta\nu_D=25$ MHz), to the large absorption at the line center ($k_{max}l=0.7$) and possibly to the broad generation line of the laser. A similar picture of dip formation should be observed also in the cesium line $6S_{1/2}(F=3)\rightarrow 6P_{3/2}$. For the D_1 line of cesium, only the dips described in Secs. 2 and 3 are possible. The nonlinear effects connected with the OP for the D lines of robidium should be analogous to effects observed in cesium.

The described resonances can be used to study the influence of small pressures of buffer gases on the collision broadening of resonance lines in the region $\Delta\nu_{coll}<\Delta\nu_D$, mixing of excited states (transition between the $6P_{3/2}$ levels influence the depth of the dip), and also to stabilize laser frequencies.

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[3]T. W. Hänsch, I. S. Shahin, and A. L. Shawlow, Phys. Rev. Lett. 27, 707 (1971).