# Resonant compensation of velocity-selective optical pumping of atoms in Doppler-free saturated absorption spectra

A. M. Akul'shin, V. L. Velichanskiĭ, R. G. Gamidov, A. Ch. Izmaĭlov, V. V. Popovichev, and V. A. Sautenkov

*P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow*

(Submitted 9 August 1990)
Zh. Eksp. Teor. Fiz. **99**, 107–114 (January 1991)

Doppler-free resonances of saturated absorption at the wavelength of the $D_2$ cesium line were investigated at various intensities and for different polarizations of counterpropagating waves. High-contrast resonances were observed in the case of a cyclic transition giving rise to the short-wavelength component of the $D_2$ line of $^{133}$Cs when the intensities of the counterpropagating waves with the left- and right-hand circular polarizations were equal. These resonances appeared because of suppression of the optical pumping of atoms in the system of the Zeeman sublevels of the ground state.

A Doppler-free resonance in a thermal ensemble of two-level nondegenerate atoms in the field of counterpropagating waves occurs as a result of a change in the degree of saturation of the absorption as the off-resonance radiation frequency (when each of the two waves interacts with its own group of atoms) changes to a resonance (so that both waves interact with the same group of atoms).[1,2]

Multilevel systems, such as atoms with a hyperfine structure, exhibit also a reduction in the number of the groups of atoms (in the velocity space) which are involved in the interaction under resonance conditions and a change in the degree of saturation, but qualitatively new effects may also be observed. Firstly, we can expect a new saturation mechanism associated with optical pumping of atoms to a long-lived and nonabsorbing state.[3-5] The saturation intensity then decreases significantly. Secondly, off-resonance excitation relaxation may occur in different groups of atoms in different channels and under resonance conditions these channels can become simultaneously accessible to a given group of atoms. It is important to note that not only the degree of saturation but also the saturation mechanism itself ("switching" on and off of a specific channel) may change.

These effects are of practical interest because they increase considerably the contrast of a Doppler-free resonance. We shall not try to provide a full classification of such effects but confine ourselves to just three specific examples of their manifestation.

1. There may be cross resonances of the $V$ type (Fig. 1a) due to combination of cyclic transitions ($A \rightarrow C$) and of transitions resulting in optical pumping of atoms from a level $A$ to a level $B$ (Ref. 4). Merging of a group of atoms in which cyclic transitions take place with a group which is optically pumped means that the former experience "switching on" of a new effective saturation mechanism, which ensures a high resonance contrast.

2. There may also be cross resonances of the $\Lambda$ type (Fig. 1b) which appear as a result of combination of transitions resulting in opposed optical pumping.[6] It is clear from the figure that the action of the pumping is opposite in each channel and when the laser frequency is tuned to a Doppler-free resonance, they balance each other out, so this saturation mechanism is "switched off."

3. The third example (Fig. 1c) is similar to the second except that counterpropagating waves are circularly polarized ($\sigma^+$ and $\sigma^-$) and that optical pumping occurs in a system of the Zeeman magnetic sublevels of one cyclic transition ($A \rightarrow C$), as described in Refs. 1, 2, and 7. If the number of sublevels of the lower level is not less than the number of them for the upper level, then optical pumping far from a resonance results in the accumulation of atoms in nonabsorbing (outer) Zeeman sublevels. When the laser radiation is tuned to a resonance, the two pumping channels cancel each other and the saturation mechanism associated with pumping is "switched off." This is manifested most strikingly for comparable intensities of the counterpropagating beams. It should be noted that earlier investigations of Doppler-free spectra of counterpropagating waves with the left- and right-hand ($\sigma^+$ and $\sigma^-$) circular polarizations have been carried out using waves with very different amplitudes.[8,9] We shall consider resonances of this type for a wide ratio of the counterpropagating wave intensities.

Our theoretical analysis will apply to one of the simplest degenerate systems, which are transitions between the states of an atom with the angular momenta $F = F' = 1/2$ (Fig. 1c). We shall analyze the behavior of two counterpropagating waves (1 and 2), with the left- and right-hand circular polarizations, traveling across a low-density (so that the collisions can be ignored) optically thin gaseous medium. We shall assume that the radiation frequency is close to the resonance frequency $\nu_0$ of a cyclic transition and that the wave intensities satisfy the condition

$$(\gamma\tau)^{-1} \ll G \ll 1, \tag{1}$$

where $\gamma$ is the homogeneous line width; $\tau$ is the effective transit time of atoms across a laser beam; $G = |E_0 d/h\gamma|^2$ is the saturation parameter; $E_0$ are the wave amplitudes; $d$ is the dipole moment of an atomic transition. The limitations on the intensity imposed by the inequalities of Eq. (1) mean that multiple absorption of light occurs during the transit time of an atom across the laser beam, whereas the probability of absorption during the lifetime of an atom in an excited state is much less than unity.

Subject to these limitations, we find that a system of equations for the density matrix[2] considered in the limit $\gamma \ll \Delta\nu_D$ ($\Delta\nu_D$ is the Doppler line width) yields the following expression for the absorption coefficient $K_1$ of a wave of intensity $I_1$ when the radiation frequency is detuned by just a

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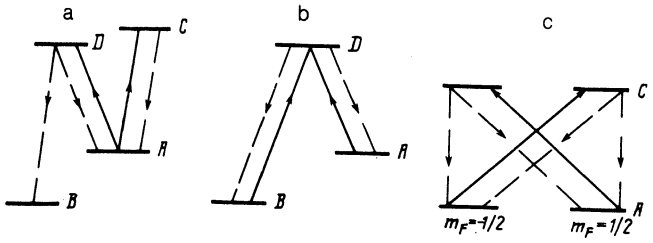


FIG. 1. Schematic representations of the energy levels of atoms which make it possible to alter the saturation mechanism by a Doppler-free resonance. The continuous arrows represent absorption and the dashed arrows represent spontaneous emission.

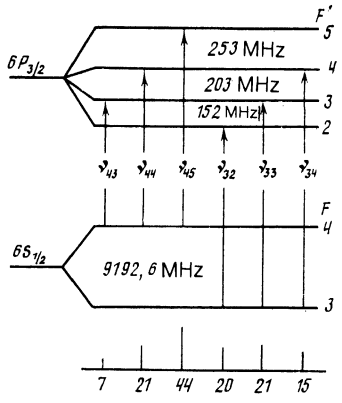


FIG. 2. Energy level scheme of the cesium atom, representing the $D_2$ line. The relative intensities of the lines representing the hyperfine transitions are given at the bottom of the figure.

small amount $\delta = \nu - \nu_0$ relative to the atomic transition frequency $(\gamma^2 + \delta^2)^{1/2} \ll \Delta\nu_D$ (Ref. 7):

$$K_1 = \frac{2K_0\gamma a}{(1+a)}[\gamma^2+4a\delta^2(1+a)^{-2}]^{-1/2}\exp\left[-\left(\frac{(1-a)\delta}{(1+a)\Delta\nu_D}\right)^2\right], \quad (2)$$

where $K_0$ is the value of $K_1$ obtained when $a = I_2/I_1 = 1$ and $\delta = 0$. It is clear from Eq. (2) that the maximum absorption and the spectral profile of the resonance are governed by the ratio of the wave intensities and not by their values. The constancy of the maximum value of $K_1$ ($\delta = 0$) on increase in the laser radiation intensity subject to the condition $a = \text{const}$ means that the resonance amplitude rises linearly on increase in the radiation power. Since for a large detuning (offset) the medium becomes effectively bleached because of the optical pumping of atoms to a nonabsorbing state, the resonance contrast should rise in accordance with the law $(I_1 + I_2)^{\alpha}$, where $\alpha > 1$.

The spectral dependence of a Doppler-free resonance differs from a Lorentzian resonance and is governed by the parameter $a$. The minimum width of the resonance of the function $K_1(\delta)$ corresponds to $a = 1$, when it becomes $K_1(\delta) \propto (\gamma^2 + \delta^2)^{-1/2}$. The total resonance width at midamplitude is then $2\times 3^{1/2}\gamma$ and is independent of the radiation intensity.

Our experiments were carried out for a more complex atomic system using the short-wavelength component [$6S_{1/2}(F=3) - 6P_{3/2}(F'=2, 3, 4)$ transition] of the $D_2$ line of the $^{133}$Cs atoms ($\lambda \approx 852$ nm) whose energy diagram is shown in Fig. 2. The hyperfine splitting of the excited state is less than the Doppler width $\Delta\nu_D \approx 400$ MHz and the frequency spacing $\Delta\nu_1 \approx 9200$ MHz between the ground-state sublevels exceeds considerably the value of $\Delta\nu_D$. The radiative width of the atomic transition is $\gamma_r \approx 5.3$ MHz.

We used the apparatus shown schematically in Fig. 3. The source of frequency-tunable radiation was an injection laser with an external cavity, described in Ref. 10. The emission of the laser was stabilized using a transmission resonance of a scanning confocal interferometer. The width of the laser emission line did not exceed 200 kHz. The output power was 2 mW. Systems of mirrors, polarizers, and quarter-wave plates were used to form two counterpropagating light beams with left- and right- circular polarizations and of $0.2\times 0.8$ cm cross section, which were combined in a cell 11 containing saturated cesium vapor. The angle between the beams meeting the cell was $\varphi \approx 7\times 10^{-3}$ rad. The radiation power was varied by calibrated neutral filters. The cell was 1 cm long and with a diameter 4 cm; it was kept at room temperature (when the cesium atom concentration was $N \approx 3\times 10^{10}$ cm$^{-3}$ and the mean free path of atoms was much greater than the cell diameter) and was surrounded by a magnetic screen which ensured that the residual field was $H \lesssim 10$ mOe. The Doppler background was removed by chopping one of the beams at a frequency of 500 Hz and recording the density of the other beam with a photodiode from which a signal was directed (after amplification and lock-in detection) to an $X$–$Y$ plotter.

The power (intensity) was varied from 5 $\mu$W ($\approx 30$ $\mu$W/cm$^2$) to 300 $\mu$W ($\approx 1.8$ mW/cm$^2$). At these intensities the saturation parameter of the transition at the frequency $\nu_{32}$ (without allowance for saturation by optical pumping) was within the range $5\times 10^{-3}$–$3\times 10^{-1}$ (Refs. 1 and 2). Since the ratio of the lifetimes $\tau_0 \approx 3\times 10^{-8}$ s of atoms in the excited state to the average transit time $\tau \approx 10^{-5}$ s through a typical beam diameter (2 mm) amounted to $(\tau_0/\tau) = 1/\gamma\tau = 3\times 10^{-3}$, the inequality $(\gamma\tau)^{-1} \lesssim G < 1$. was satisfied in our experiments.

Figure 4 shows Doppler-free resonances observed for the short-wavelength component of the $D_2$ line recorded when the power (intensity) of each of the counterpropagating waves was 300 $\mu$W ($\approx 1.8$ mW/cm$^2$) in Fig. 4a and 5 $\mu$W ($\approx 30$ $\mu$W/cm$^2$) in Fig. 4b. We identified the intrinsic $\nu_{FF'}$, and cross $(\nu_{32} + \nu_{33})/2$, and $(\nu_{32} + \nu_{34})/2$, and $(\nu_{33} + \nu_{34})/2$ resonances[11] at the bottom of this figure. In the system under consideration a nonlinear change in the absorption could be due to pumping of atoms to: 1) an excited state: 2) the hyperfine sublevel $F = 4$; 3) the nonabsorbing magnetic sublevels of the state $F = 3$. Since the intensities used in our experiments ensured that the inequality $G < 1$ was satisfied, we could ignore the first of these mechanisms. The second mechanism was observed in its pure form for the resonance at the frequency $\nu_{34}$ and the third for the resonance at $\nu_{32}$.

The signs of the resonances agreed with these two mechanisms. In fact, in the case of the lowest-frequency resonance when the laser frequency was tuned exactly to this resonance, the process of optical pumping in the system of the magnetic sublevels of the $F = 3$ state was compensated

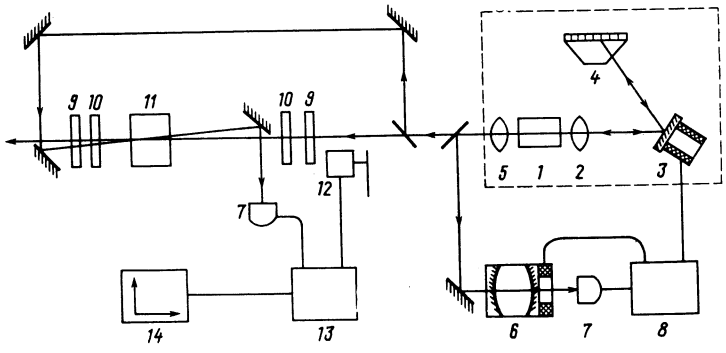


FIG. 3. Schematic diagram of the apparatus: 1) injection laser; 2) matching microobjective; 3) mirror on a piezoelectric ceramic substrate; 4) holographic selector; 5) exit objective; 6) confocal scanning interferometer; 7) photodiode; 8) automatic frequency control unit; 9) polarizer; 10) quarter-wave plate; 11) cell with the $^{133}$Cs vapor; 12) chopper; 13) resonant amplifier and lock-in detector; 14) $X$-$Y$ plotter.

by the opposite effect of the counterpropagating laser beams and the absorption increased. In the case of the resonance at the frequency $\nu_{34}$ the pumping of the atoms to the $F=4$ level was additive for each of the two waves when the laser radiation was tuned exactly to the resonance, so that the degree of bleaching increased and the absorption fell. In all the other resonances both mechanisms were active, so that a qualitative interpretation of their sign was difficult.

A numerical calculation, carried out using a theory of Ref. 11 for the same polarizations ($\sigma^+$ and $\sigma^-$), yielded the amplitudes and signs of the resonances given in the lower part of Fig. 4. It should be noted that the calculations were carried out on the assumption that the saturation was weak, i.e., the intensities were such that the probability of absorption in a saturating beam was less than 1 even during the whole of the transit time ($G\gamma\tau < 1$) and subject to the condition that the power of a probe beam $P_p$ was much less than the power of the saturating beam $P_s$. It is clear from Fig. 4 that the signs of the resonances represented by the experimental curves were in agreement with the theoretical results obtained for weak fields in Ref. 11.

Moreover, the relative amplitudes in the spectrum in Fig. 4b were in satisfactory agreement with the predictions of the theory of Ref. 11. A reduction in the power of the probe wave by a factor of 10 (from $P_p \approx P_s \approx 5$ $\mu$W to $p_p \approx 0.1 P_s$) ensured an even better agreement between the experimental and theoretical spectra. The signs of the resonances were not affected by an increase in the intensities of the counterpropagating waves or by equalization of these intensities. Out of all the cross resonances only the one at the lowest frequency was formed as a result of a combination of the transitions and each of these admitted could be due to the third of the saturation mechanisms listed above. Clearly, this was the reason for the observed sign. However, an increase in the wave intensities activated a second electron-liberating channel $F=3 \rightarrow F'=3 \rightarrow F=4$ and the amplitude of this resonance [at a frequency $(\nu_{32}+\nu_{33})/2$] was lower than the amplitude of the resonance at the frequency $\nu_{32}$.

The conditions for the formation of the latter (lowest-frequency) resonance at $\nu_{32}$ were closest to the model assumed in obtaining the result represented by Eq. (2). The dependences of the amplitude $A$ and of the width $\Gamma$ at mid-amplitude of the Doppler-free resonances at the frequencies $\nu_{32}$ and $(\nu_{32}+\nu_{33})/2$ on the power $P=P_s \approx P_p$ of the counterpropagating waves were determined (Figs. 5 and 6). The dependences for the cross resonance were of the conventional form: an increase in the intensity increased the resonance

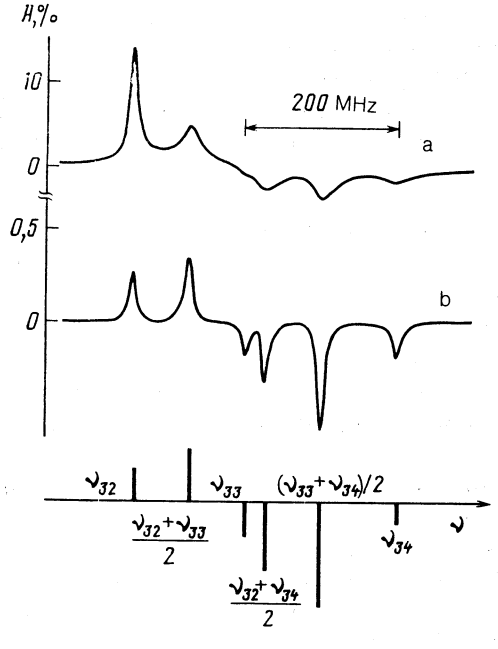


FIG. 4. Saturated absorption spectra for the short-wavelength component [$6S_{1/2}(F=3)$–$6P_{3/2}(F'=2, 3, 4)$ transition] of the $D_2$ line of cesium ($\lambda \approx 852$ nm) obtained when the power (intensity) of each of the counterpropagating circularly polarized ($\sigma^+$ and $\sigma^-$) waves was: a) 300 $\mu$W ($\approx 1.8$ mW/cm$^2$); b) 5 $\mu$W ($\approx 30$ $\mu$W/cm$^2$). One division along the ordinate represents 1% transmission of the probe wave.

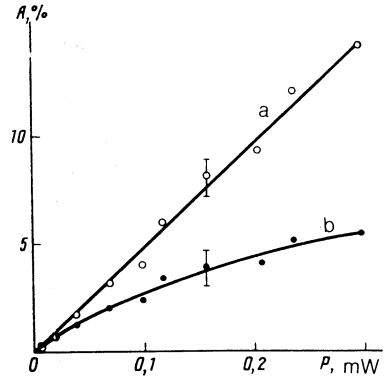


FIG. 5. Dependences of the amplitude $A$ of Doppler-free resonances at the frequencies $\nu_{32}$ (a) and $(\nu_{32}+\nu_{33})/2$ (b) on the power $P=P_p \approx P_s$ of the counterpropagating waves with the $\sigma^+$ and $\sigma^-$ polarizations. One division along the ordinate represents 1% transmission of the probe wave.

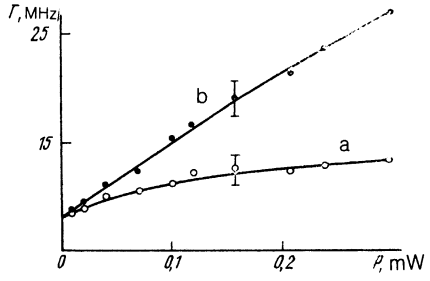


FIG. 6. Dependence of the width Γ at midamplitude on the power $P = P_p \approx P_s$ of the counterpropagating waves with the $\sigma^+$ and $\sigma^-$ polarizations recorded for a Doppler-free resonance at the frequency $\nu_{32}$ (a) and for the adjacent cross resonance at $(\nu_{32} + \nu_{33})/2$ (b).

width and the rise of the resonance amplitude rapidly reached saturation. All the other resonances behaved similarly (apart from the sign), but this was not true of the lowest-frequency resonance at $\nu_{32}$. The behavior of this lowest-frequency resonance was unusual: the amplitude rose linearly on increase in the intensity, whereas the resonance width increased slowly from $\gamma$ to $1.5\gamma$ (after allowance for the contribution made to the increase in the angle between the counterpropagating waves amounting to $\gamma \approx 8$ MHz).

One should mention here a disagreement with the theory according to which the resonance width should remain constant in this region and its steady-state value should be considerably greater ($\approx 3.4\gamma$). In our opinion this discrepancy was due to the complexity of the real system ($F = 3 \rightarrow F' = 2$ transition involved the interaction of 12 sublevels) compared with the model Fig. 1c, and also because the condition (1) was not satisfied rigorously particularly at low intensities [the inequality $(\gamma\tau)^{-1} \lesssim G < 1$ was satisfied in our experiments].

We checked the role of the polarization and of the factor $a$, and studied the behavior of the resonance contrast at the frequency $\nu_{32}$ by recording the spectra without removal of the Doppler background; this was done for different polarizations and different intensities of the counterpropagating waves (Fig. 7). Clearly, an increase in the probe beam intensity reduced the contrast of all the resonances, with the exception of the contrast of the resonance at $\nu_{32}$ ($\sigma^+$, $\sigma^-$), which increased. When the polarizations were linear, all the special features of the behavior of the resonance at $\nu_{32}$ were suppressed. This was to be expected because the selective (in respect of the velocity of the atoms) resonant compensation of the optical pumping did not occur in the case of the linearly polarized waves. When the detuning was large, each of the waves "chased" the atoms away to the states with $F = 3$ and $m_F = \pm 3$, i.e., it ensured alignment in each of the groups of atoms and the tuning to the resonance simply altered the degree of alignment. In the case of the $\sigma^+$ and $\sigma^-$ polarizations of interest to us we found that in each of the groups of atoms far from a resonance the orientations with opposite signs were induced, whereas in the case of an exact tuning to a resonance the orienting effects of the counterpropagating waves were balanced out exactly.

It was reported in Ref. 8 that the sign of a cross resonance at a frequency $(\nu_{32} + \nu_{33})/2$ was opposite to that observed in our study. A theoretical calculation reported in Ref. 8 also predicted a negative sign of this resonance,

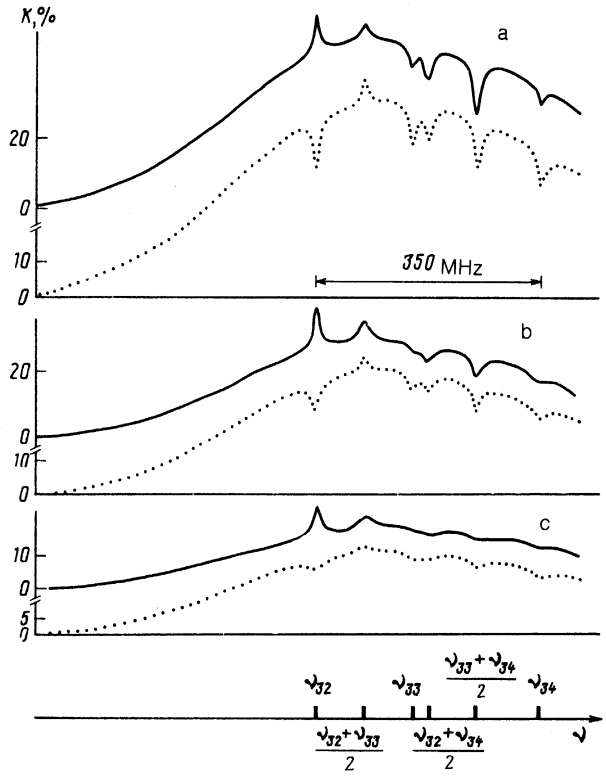


FIG. 7. Saturated absorption spectrum (recorded without suppression of the Doppler background) obtained for the circular ($\sigma^+$ and $\sigma^-$) polarizations (continuous curves) and for the same linear ($\pi$, $\pi$) polarizations (dashed curves) of the counterpropagating waves: a) $P_p \approx 0.1 P_s$; b) $P_p \approx 0.3 P_s$; c) $P_p \approx P_s$; $P_p$ is the probe wave power and $P_s \approx 140\,\mu$W is the power of the saturating wave. One division along the ordinate represents 1% of the absorption of the probe wave.

whereas the theory developed in Ref. 11 predicted a positive sign. Our experimental investigations in a magnetic field of $H = 0.01$–$10$ Oe intensity showed that a cross resonance should be positive in the case when the longitudinal component $H_\parallel$ was much greater than the transverse component $H_\parallel \gg H_\perp$, and should be negative if $H_\parallel \ll H_\perp$. For $H_\parallel \gtrsim H_\perp$ ($H \approx 0.1$–$1$ Oe, which was typical of the laboratory conditions) the positive sign should change to negative on increase in the intensities of the counterpropagating waves. For example, in a magnetic field with the components $H_\parallel \approx 1$ Oe and $H_\perp \approx 0.3$ Oe the sign of the resonance was positive for $I_p \approx 0.025$ mW/cm$^2$ and $I_s \approx 0.6$ mW/cm$^2$, but it was negative for $I_s \approx 2.5$ mW/cm$^2$. When the cell was inside the magnetic screen, it was found that in the range of intensities $I_p \approx I_s \approx 0.02$–$6$ mW/cm$^2$ the sign of the cross resonance in our experiments was positive and it corresponded to the case when $H_\parallel \gg H_\perp$.

Clearly, the authors of Ref. 8 did not monitor the transverse component of the field.

We thus found that the $D_2$ lines of alkali elements could be used to induce high-contrast narrow resonances for the same intensities of the probe and saturating waves and these resonances should be suitable for the stabilization of the frequencies of injection lasers,[12] particularly when a cell is placed inside the resonator. The advantage of such resonances in an application of this kind is a weak dependence of their profiles on the intensity of laser radiation, as pointed out in Ref. 7.

We explained the theoretical results using two models: one of them[7] simplifies greatly the energy spectrum, but is suitable when the saturation parameters are large, whereas the other[11] allows for the real structure of the atomic system, but applies so far only to the low values of the saturation parameters. Generalization of the theory of Ref. 7 to the case of large saturation parameters should make it possible to carry out a more detailed comparison with the experimental data.

An attractive feature in the system for the excitation of atoms with light characterized by $\sigma^+$ and $\sigma^-$ circular polarizations and the same intensity of the counterpropagating beams is the feasibility of cyclic excitation of atoms (due to the absence of bleaching of the gaseous medium at these frequencies), which is of interest in connection with the development of a three-dimensional optical trap for neutral atoms.[13,14]

The authors are grateful to V. I. Yudin for valuable discussions and for supplying the results of calculations based on Ref. 11, and to V. A. Alekseev for stimulating discussions.

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Translated by A. Tybulewicz