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Observation of the intra-Doppler Zeeman splitting of the D1 line of atomic cesium in low magnetic fields by the method of saturated absorption spectroscopy

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A simple model is proposed in the paper, which allows quantitative description of experiments on observation of the Zeeman effect on the D1 line in atomic cesium vapors by the method of saturated absorption spectroscopy (SAS). It is shown that simple exclusion of the contribution of the zero velocity group to the absorption spectrum is not sufficient for interpretation of the SAS results of atomic cesium in the region of the D1 line. The experiments presented in the paper, illustrating the proposed model, were carried out in a low magnetic field, when the Zeeman splitting of atomic multiplets was significantly less than the Doppler broadening.

Keywords: alkali metal vapor spectroscopy, cesium D1 line, saturated absorption spectroscopy, unresolved Zeeman structure.

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1. Introduction

Laser saturated absorption spectroscopy (SAS) in opposite beams (Doppler-free laser spectroscopy) is the effective method to observe and study the atomic energy structure under the conditions of considerable Doppler widening [1–8]. Wide application of tunable semiconductor lasers considerably simplifies the SAS experiments, however, their interpretation is not always evident in virtue of the nonlinear nature of the SAS method as such [1,2,9]. In the proposed paper, using the SAS method, Zeeman splitting is observed in the components of the cesium (Cs) D1 line in small magnetic fields in the Voigt geometry, when it is substantially smaller than the Doppler width of the components, and its direct observation is not possible. Using the proposed simple model, it was possible to quantitatively interpret the obtained spectra, which have a rich structure related to the difference in the values and signs of g-factors of the main (g_g) and excited (g_e) multiplets of Cs atom shaping the D1 lines.

The work is structured as follows. The second section describes the SAS method and the experimental setup. Using the example of the spectra of atomic vapors of cesium we obtained using the SAS method in the weak magnetic field, the inadequacy of the simple exclusion of the contribution from the atoms of the zero speed group for interpretation of the specified spectra is demonstrated. The third section describes the necessary data on the relaxation mechanisms in the atomic system and on the modeling of atomic spectra. The fourth section specifies the theoretical model making it possible to quantitatively interpret the magnetic behavior of the spectra. The fifth section describes the experiment and provides the necessary comments. The results of the paper are briefly summarized in the Conclusion.

2. Saturated absorption method. Experimental setup

Let us explain the SAS method [2] and also describe the experimental setup used in our paper in Fig. 1, *a*. A weak probing beam (it spreads from the left to the right) is produced by splitting of the tunable laser beam 1 by a beam-splitting plate 2 and weakening of its intensity to the value of $\sim 1\,\mu$W. Having passed cell 4 with Cs vapors, the probing beam enters photodetector 5, the output signal of which is recorded by oscilloscope 6. When the frequency of the tunable laser is scanned in the area of the atomic vapors absorption line (we worked in the spectral area of line D1 of cesium vapors $\lambda = 894.593$ nm) the oscilloscope screen shows an absorption spectrum of the atomic system in the channel of the weak probing beam, which (the spectrum) in our experiments did not depend on the intensity of the probing beam (nonperturbation of the probing beam).

In the SAS method, except for a weak probing beam, a relatively strong pumping beam affects the studied system of the atomic vapors, which spreads towards the probing beam [2,10] and does not enter the photodetector (Fig. 1, *a*). In our setup this beam was shaped by mirror 7 and semitransparent plate 8. The capacity of the pumping beam usually was $\sim$5 mW and could change with the help of attenuator 9. The probing beam and the pumping beam had the diameter of ~ 3 mm and were usually polarized perpendicularly to the magnetic field (along axis X of the laboratory system of coordinates, Fig. 1, *a*). The

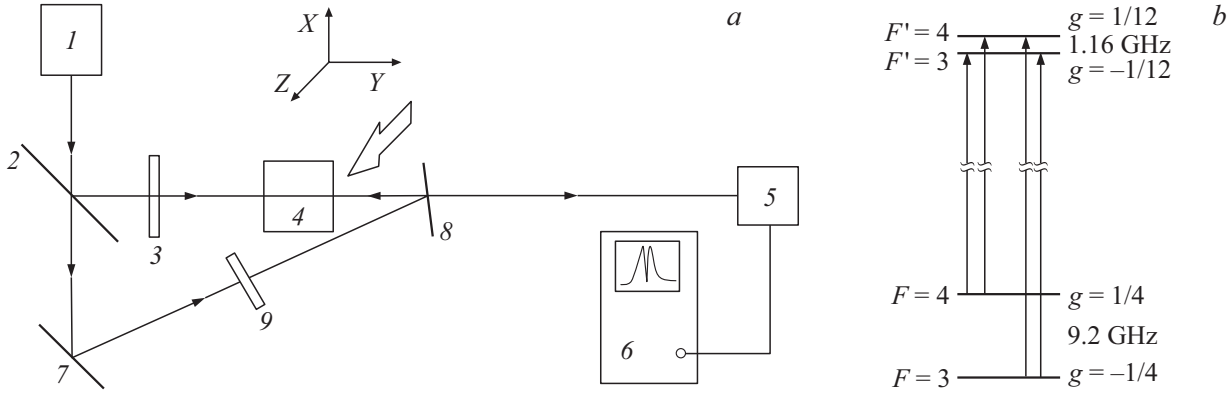

Figure 1. (*a*) Diagram of the experimental setup. 1 — tunable laser, 2 — beam-splitting plate, 3 — attenuator, 4 — cell with Cs pairs, 5 — photodetector, 6 — oscilloscope, 7 — mirror, 8 — semi-transparent plate, 9 — attenuator. (*b*) Transitions shaping a D1 line of cesium. A full moment (on the left) and *g*-factor (on the right) are shown near each multiplet.

experiments were carried out at temperature of 22 °C, the cell length was 50 mm.

The SAS method consists in registration of the change in the absorption spectrum in the atomic system, observed in the channel of the weak probing beam, when the pumping beam is activated (deactivated) (Fig. 1, *a*). Let us designate using Ω and ω accordingly the frequency of the atomic transition that is not displaced by the Doppler effect and the frequency of the laser beams. As it is known, the atoms of the speed group **v**, which are present in the field of the electromagnetic wave with the wave vector of $\mathbf{k} = (0, k, 0)$, $k \equiv \omega/c$, (c — light velocity), experience excitation in the displaced frequency $\omega + (\mathbf{k}, \mathbf{v}) = \omega + kv_y$ (Doppler effect). Accordingly, in the general case the probing and pumping beams (which have the opposite signs of the wave vectors) are resonant for the atoms of different speed groups: the probing beam is resonant to the atoms of the speed group $v_y^{pr} = [\Omega - \omega]/k$, and the pumping one — of the group $v_y^{pu} = [\omega - \Omega]/k$. Therefore, in the general case the activation (deactivation) of the pumping beam will not affect anyhow the transmission coefficient of the probing beam — even if the pumping beam fully elucidate the atoms of the speed group $v_y^{pu} = [\omega - \Omega]/k$, and the state of the probed atoms of the speed group $[\Omega - \omega]/k$ will not change. The exception is the situation when the probed and pumped groups coincide, which happens at $\omega = \Omega$. In this case the pumping beam elucidates the probed zero speed group of atoms ($v_y = 0$), therefore, in the center of the absorption line of atomic vapors observed in the channel of the probing beam, inclusion of the pumping beam causes appearance of a dip (Fig. 2, *a*). If the pumping beam is not very powerful, the dip width (in our case ~ 10 MHz) is determined by homogeneous width δ of atomic transitions and usually is substantially less than the Doppler width $\Delta \equiv kv_T$, $v_T \equiv \sqrt{\frac{2k_BT}{m}}$, where T — temperature, k_B — Boltzmann constant, m — atomic weight (Fig. 2, *a*). Observation and characterization of this dip are the content of the SAS method, which makes it possible to obtain information on the studied atomic system at substantially higher spectral resolution compared to the Doppler width Δ [4]. In the proposed paper the SAS method will be used to observe the Zeeman splitting of multiplets in the atomic cesium D1 line in small magnetic fields. The observation of such splitting by the SAS method in the electric field was described in [3,11]. Use of the SAS method to record the Zeeman effect in CH_4 molecules is described in [3,12]. We use the SAS method to observe the Zeeman splitting in atomic pairs of cesium [13], and the main content of our paper consists in the simple quantitative interpretation of these experiments.

In the simplest consideration the effect that the SAS method is based on may be imagined as follows. Let us designate via $A(\nu)$ the absorption spectrum of the „atom at rest“ ($\nu \equiv \Omega - \omega$ — optical detuning). Then the absorption spectrum of the atomic system $a_0(\nu)$ widened with Doppler effect with the deactivated pumping beam may be expressed with the help of Maxwell distribution $\rho_M(v)$ as

$$a_0(\nu) = \int \rho_M(v) A(\nu - kv) dv, \qquad (1)$$

$$\rho_M(v) \equiv N \exp\left(-\frac{v^2}{v_T^2}\right) \frac{1}{\sqrt{\pi} v_T}, \qquad v_T \equiv \sqrt{\frac{2k_BT}{m}}.$$

Here N — total quantity of atoms in the probing beam. As the pumping beam is activated, the contribution of the speed group $v = 0$ leaves the absorption spectrum as this group is elucidated by the pumping beam. „The volume“ δv of this speed group in the space of speeds may be estimated as $\delta v \sim \delta/k$ (where δ — homogeneous width of the atomic transition), after which the absorption spectrum in the channel of the probing beam recorded with the activated pumping beam may be presented as

$$a(\nu) = \int \rho_M(v) A(\nu - kv) dv - \frac{\xi\delta}{k} \rho_M(0) A(\nu), \qquad (2)$$

where ξ — adjustable parameter, besides, in the typical case, when the pumping beam elucidates the resonant

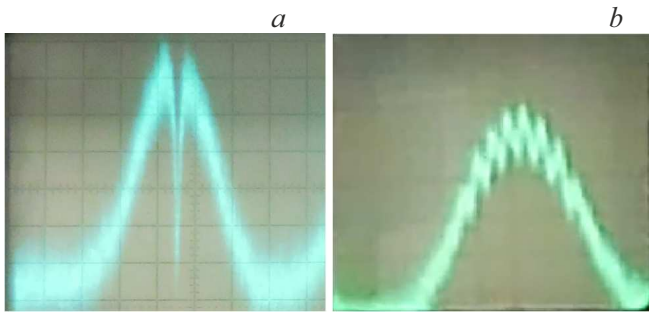

Figure 2. (a) Dip in the absorption spectrum of Cs vapors recorded at the setup of Fig. 1, with the activated pumping beam. (b) Dip splitting in transverse magnetic field $B \approx 100$ gauss.

speed group practically fully, but does not result yet in the substantial increase of the "volume" of this group compared to the value δ/k introduced above, parameter $\xi \sim 1$ (Fig. 2, a).

As we will see below, the described pattern of dip formation in the spectrum obtained using the SAS method, needs substantial correction when the atomic system behavior is analyzed in the static magnetic field. The magnetic field **B** in our experiments was applied perpendicularly to both beams and was so low ($B \sim 100$ gauss) that the absorption spectrum recorded with the deactivated pumping beam hardly depended on its value B. This meant that the Zeeman splitting of atomic multiplets was substantially smaller than the heterogeneous Doppler width Δ. As for the dip recorded using the SAS method, it found the confidently observed Zeeman splitting (Fig. 2, b), which substantially depended on the ratio of full moments of the main and excited multiplets of cesium D1 line components (compare the top and bottom spectrum in Fig. 3). Below a simple theory will be built, making it possible to quantitatively describe the observed magnetic behavior of the spectrum of absorption of atomic cesium vapors registered using the SAS method.

3. Hyperfine pumping and absorption spectrum of „atom at rest"

Prior to changing to building the above theory, we will specify the mechanism of depletion of populations of atomic states of the main and excited resonance multiplets, which we will consider the main one in interpretation of our experiments. Let us assume that the pumping beam saturates the transition $F = 4 \to F' = 3$ of cesium line D1 (Fig. 1, b). We will believe that initially the states of resonant ($F = 4$) and non-resonant ($F = 3$) main multiplets are equally populated, and populations of excited multiplets are equal to zero. Pumping of transition $F = 4 \to F' = 3$ populates the excited resonant multiplet $F' = 3$, from where the fast radiation decomposition into both main multiplets $F = 4$ and $F = 3$ occurs. Since pumping empties only multiplet $F = 4$, multiplet $F = 3$ is effectively populated, and the populations of the states of the resonant main multiplet $F = 4$ are depleted. This process is called hyperfine pumping [14,15], and we will assume it is the main one in our consideration.

Let us designate the atomic states of the main (excited) resonant multiplet as $|g, M\rangle$ ($|e, M\rangle$), where $M = -F, \ldots, F$ ($M = -F', \ldots, F'$) is a projection of atomic moment to the direction of the magnetic field. Then the energies of the states of the main (excited) multiplet may be expressed using Bohr magneton μ as: $E^g_M = \omega_{L2} M$, ($E^e_{M'} = \Omega + \omega_{L1} M'$), where $\omega_{L2} = g_g \mu B/\hbar$ ($\omega_{L1} = g_e \mu B/\hbar$) – Larmor's frequencies of the main (excited) multiplet. Using these definitions, the absorption spectrum of „atom at rest" introduced above may be presented in the form of (let us omit the dimension coefficient that are not significant for us at the moment)

$$A(v) = $$

$$= \sum_{MM'} \mathcal{L}(\omega_{L2} M' - \omega_{L1} M - v)\ [\sigma_{gM} - \sigma_{eM'}] |\langle eM'|D|gM\rangle|^2,$$

$$\mathcal{L}(x) \equiv \frac{1}{\pi} \frac{\delta}{\delta^2 + x^2}, \quad D = S_x \sin\phi + S_z \cos\phi. \qquad (3)$$

Here σ_{gM} ($\sigma_{eM'}$) — normalized per unit of population of the states of the main (excited) multiplet, ϕ — azimuth of linear polarization of the probing beam, and matrix elements of vector operators $S_{x,z}$ between the states with the specified projection of the moment are known in clear in an explicit form [16]. With the deactivated pumping beam $\sigma_{gM} = 1/[2F + 1 + 2f + 1] \equiv s$, $\sigma_{eM'} = 0$, where f — full moment of the non-resonant main multiplet. The direct substitution of formula (3) into expression (2) makes it possible to describe the dip observed with the help of SAS method (Fig. 2, a) in the zero magnetic field ($\omega_{L1} = \omega_{L2} = 0$). However, the attempt to describe so the splitting of the dip caused by the transverse magnetic field (i.e. at $\omega_{L1,2} \neq 0$) demonstrates the noticeable mismatch in the estimated and theoretical spectra (Fig. 3), which indicates the need for more detailed consideration of the atomic system saturation with the presence of the magnetic field.

4. Theory of saturation of atomic vapors in low magnetic field

Let us first consider our atomic system with the deactivated pumping beam. The speed groups $v_{MM'}$[1], defines as follows, as parallel (antiparallel) to the probing (pumping) beam:

$$v_{M'M} = \frac{\omega - E^e_{M'} + E^g_M}{k}. \qquad (4)$$

Atoms of such speed groups (in the general case the quantity of these groups $(2F + 1)(2F' + 1)$) form the

[1] are resonant for the probing laser beam with frequency of ω . Below it means y-component of the speed of atoms.

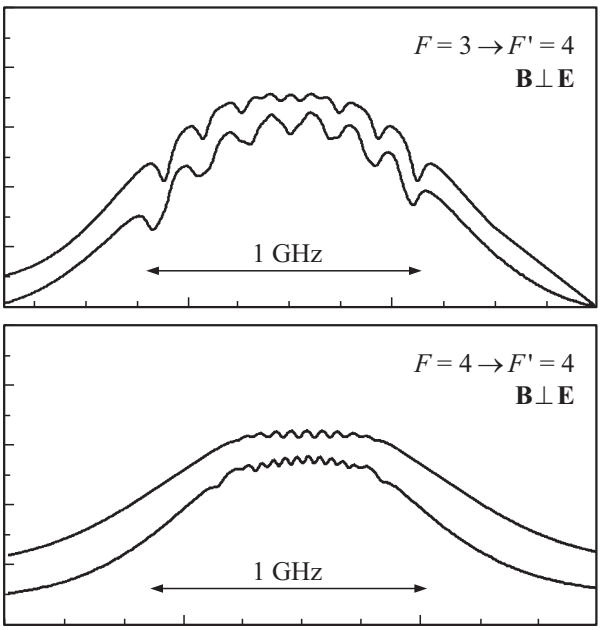

Figure 3. Observation of Zeeman splitting of D1 line components of the cesium absorption spectrum by SAS method in the magnetic field $B \approx 100$ gauss: transition $F = 3 \to F' = 4$ (top figure), $F = 4 \to F' = 4$ (bottom figure). The spectra are shown in arbitrary units. Besides, the spectra are also given, which are obtained by subtraction of the zero speed group using formula (2) (curves displaced upwards). The horizontal two-directional arrow shows a frequency scale.

observed absorption in the channel of the probing beam. If populations of states, from which the transitions in these groups happen (let us call these states start ones), are changed, the registered absorption will change as well.

Let us now activate the pumping beam. Let us designate, using $\tilde{v}_{L'L}$, the speed groups that this beam saturates in resonant manner. They differ from $v_{M'M}$ (4) by the sign of the factor k (i.e. $k \to -k$):

$$\tilde{v}_{L'L} = \frac{E^e_{L'} - E^g_L - \omega}{k}. \tag{5}$$

For the pumping to affect the atoms of the speed group $v_{M'M}$ (4), it is necessary that a group is found among groups $\tilde{v}_{L'L}$ (5) that coincides with $v_{M'M}$, i.e. such L and L' are found that $\tilde{v}_{L'L} = v_{M'M}$ and, accordingly, $E^e_{L'} - E^g_L - \omega = \omega - E^e_{M'} + E^g_M$. Besides, it is necessary that in the speed group $\tilde{v}_{L'L}$ the pumping beam depletes the same state $|gM\rangle$, which is a start one for the probing beam, i.e. it must be: $E^g_L = E^g_M$. The compliance with these two requirements causes the following condition for the frequency of the beams ω:

$$\omega = \frac{E^e_{L'} + E^e_{M'}}{2} - E^g_M. \tag{6}$$

If this condition is met, the pumping beam effectively saturates the atoms of the speed group $v_{M'M}$ (these atoms are transferred to the states of the non-resonant main multiplet due to the above mechanism of hyperfine pumping), and their contribution to the absorption of the probing beam „drops out“, i.e. becomes negligibly small. Besides, it is necessary that the corresponding transition for the pumping beam was resolved. Therefore, frequencies $\omega_{L'M'M}$, at which the activation of the pumping beam causes dips are determined by expression (6) and the following condition:

$$|\langle gM|S_x \sin\theta + S_z \cos\theta|eL'\rangle|^2 \neq 0. \tag{7}$$

Here θ — azimuth of linear polarization of the pumping beam (it may not coincide with azimuth ϕ of the probing beam).

When the frequency of the probing beam $\omega = \omega_{L'M'M}$, then, as you can see it from (4) and (6), the speed group is probed $v_{M'M} = [E^e_{L'} - E^e_{M'}]/2k$. The number of atoms in this group is $\sim s\rho_M([E^e_{L'} - E^e_{M'}]/2k)\delta v$. Having this mind, to change the absorption spectrum along the probing beam $\Delta a(\nu)$, arising in the activation of the pumping beam, the following expression may be written:

$$\Delta a(\nu) = s \sum_{L'M'M} \mathcal{L}\left(\omega_{L'M'M} - \omega\right) \times |\langle eM'|S_x \sin\phi + S_z \cos\phi|gM\rangle|^2 \times J\left(|\langle gM|S_x \sin\theta + S_z \cos\theta|eL'\rangle|^2\right)\rho_M\left(\frac{E^e_{L'} - E^e_{M'}}{2k}\right)\delta v. \tag{8}$$

Here the function $J(x)$ implements condition (7), i.e. such that $J(0) = 0$ and $J(x \neq 0) = 1$. The value δv was estimated by us above as $\delta v \sim \delta/k$.

The results of adjustment of the experimentally produced spectra of absorption of the cesium vapors in the transverse magnetic field using SAS method (Fig. 1) with the help of formula (8) are given in Fig. 4. The test function had the view of $a(\nu) = a_0(\nu) - \xi\Delta a(\nu)$, where $a_0(\nu)$ was calculated using expressions (1) and (3):

$$a_0(\nu) = s \int d\nu \rho_M(v) \sum_{MM'} \mathcal{L}(\omega_{L2}M' - \omega_{L1}M - \nu + kv) \times |\langle eM'|S_x \sin\phi + S_z \cos\phi|gM\rangle|^2 \approx (\text{at}\Delta \gg \delta) \approx \frac{s}{k} \sum_{MM'} \rho_M\left(\frac{\nu - \omega_{L2}M' + \omega_{L1}M}{k}\right) \times |\langle eM'|S_x \sin\phi + S_z \cos\phi|gM\rangle|^2. \tag{9}$$

Practically all the parameters important for us that are included in (8) and (9) are known from literature [17]. The developed formulae for $\Delta a(\nu)$ and the required matrix elements are given in Appendix.

5. Experiment

The above discussions and estimates presume that the dips in the absorption spectrum registered using SAS

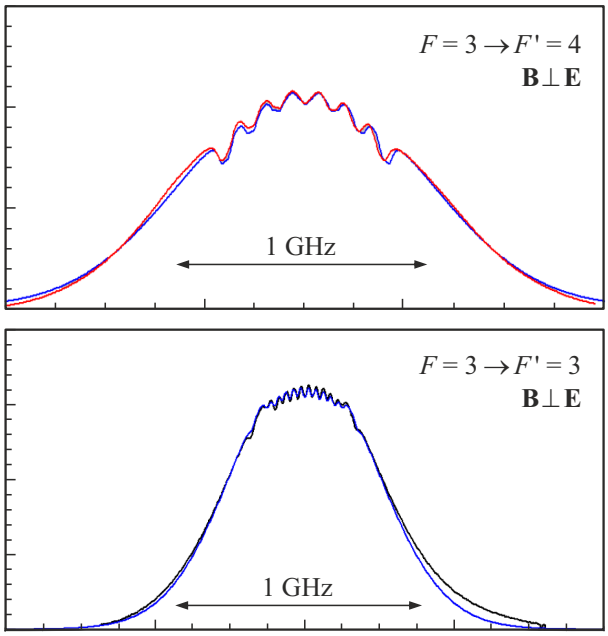

Figure 4. Observation of Zeeman splitting of D1 line components of the cesium absorption spectrum by SAS method: transition $F = 3 \rightarrow F' = 4$ top figure ($F = 3 \rightarrow F' = 3$ bottom figure). $B \approx 100$ gauss. The spectra are shown in arbitrary units. The results are shown by adjustment of experimental spectra using formula (8) (smooth curves). The horizontal two-directional arrow shows a frequency scale.

method arise as a result of the practically complete depletion of the start states of the transitions providing for the absorption of the weak probing beam. The intensity of the probing and pumping beams required to achieve such mode in our experiments was set in the following manner. At the first stage the magnetic field was deactivated, the pumping beam was closed, and the intensity of the probing beam was selected so small ($\sim 1\,\mu$W at the diameter of the beam ~ 3 mm) that the spectrum of absorption of the cell with the cesium vapors achieved by scanning of frequency ω of the tunable laser 1 (Fig. 1) stopped depending on the intensity. When the pumping beam was opened, a dip appeared in the center of the spectrum (Fig. 2, *a*), and the intensity of the pumping beam was such that the dip amplitude was practically full (Fig. 2, *a*), and its width insignificantly exceeded its value obtained at the lower intensity of pumping. Such mode was easy to achieve at intensities of pumping $\sim 1-5$ mW, after which the achieved spectra with the dip were adjusted with the help of functions $\Delta a(\nu)$ (8) and $a_0(\nu)$ (9) at $\omega_{L1,2} = 0$ and $\xi \sim 1$. Then the transverse magnetic field was activated, besides, the pattern of dip splitting was reproduced well by formulae (8), (9) (Fig. 4). The probing and pumping beams were linearly polarized perpendicularly to the magnetic field ($\theta = \phi = \pi/2$) — at the same time the spectra at $F = F'$ (Fig. 4, bottom figure) turned out most effective.

The rich structure of the spectra in Fig. 4 was related to the difference in g-factors of the main and excited multiplets of cesium ($|g_g| = 1/4$ and $|g_e| = 1/12$), as a result of which three frequency „stairs“ shifted towards each other with a pitch of $M[\omega_{L1} - \omega_{L2}] = M\mu B[g_e - g_g]/\hbar, M = -F, \ldots, F$ (see (P1)). If probed multiplets have different full moments (Fig. 4, top figure), then due to the difference of signs of g-factors the specified „staircase“ turns out to be more rarefied than in the case of match of full moments (Fig. 4, bottom figure), when the signs of g-factors match.

6. Conclusion

The paper describes the experiments to observe Zeeman splitting of multiplets in the D1 line of atomic cesium by SAS method in small magnetic fields, when the specified splitting is substantially lower than the Doppler width of the line, and a simple model is proposed for their quantitative description. It was shown that the spectra registered by SAS method are not produced by simple exclusion of the contribution of the zero speed group, but require more consistent consideration of depletion of atomic states with the pumping beam.

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Conflict of interest

The author declares that he has no conflict of interest.

Appendix

Let us show the expressions used to build theoretical spectra of Fig. 4 in the developed form. In the formulae below the matrix elements of operator S_x included in expression (8) at $F' = F \pm 1, F$ are expressed through matrix elements of operator $S_+ \equiv S_x + \imath S_y$ [16]. Besides,

since $\omega_{L1,2} \ll \Delta$, we agreed $\rho_M\left(\frac{E^e_{L'}-E^e_{M'}}{2k}\right) \equiv \rho_M(0)$:

$$\Delta a(\nu) = \rho_M(0)\,\frac{\delta}{k}\sum_{l=-1}^{1}\sum_{M=-F}^{F}\left\{\frac{\sin^2\phi}{4}\right.$$
$$\times\,\mathcal{L}\left(\nu + M[\omega_{L1}-\omega_{L2}] + \omega_{L1}\frac{l+1}{2}\right)\left|\langle F', M+1|S_+|FM\rangle\right|^2$$
$$+\,\frac{\sin^2\phi}{4}\,\mathcal{L}\left(\nu + M[\omega_{L1}-\omega_{L2}] + \omega_{L1}\frac{l-1}{2}\right)$$
$$\times\left|\langle F, M|S_+|F', M-1\rangle\right|^2 + \cos^2\phi$$
$$\left.\times\,\mathcal{L}\left(\nu+M[\omega_{L1}-\omega_{L2}]+\omega_{L1}\frac{l}{2}\right)\left|\langle F', M|S_z|FM\rangle\right|^2\right\}J(X_{Ml}), \quad \text{(A1)}$$

where

$$X_{M,l} \equiv \frac{\delta_{l,1}\sin^2\theta}{4}\left|\langle F', M+1|S_+|FM\rangle\right|^2$$
$$+\,\frac{\delta_{l,-1}\sin^2\theta}{4}\left|\langle F, M|S_+|F'M-1\rangle\right|^2$$
$$+\,\delta_{l,0}\cos^2\theta\left|\langle F', M|S_z|FM\rangle\right|^2, \quad \text{(A2)}$$

and

$$\langle F, M+1|S_+|F-1, M\rangle = \sqrt{\frac{(F+M)(F+M+1)}{2}}, \quad \text{(A3)}$$
$$\langle F-1, M+1|S_+|F, M\rangle = -\sqrt{\frac{(F-M)(F-M-1)}{2}},$$
$$\langle F, M|S_z|F-1, M\rangle = \langle F-1, M|S_z|F, M\rangle = -\sqrt{\frac{F^2-M^2}{2}},$$
$$\langle F, M+1|S_+|F, M\rangle = \sqrt{F(F+1)-(M+1)M}, \quad \text{(A4)}$$
$$\langle F, M|S_z|F, M\rangle = M.$$

In calculations the function $J(x)$, implementing condition (7), was taken as $J(x) = \frac{x^2}{\delta_1^2+x^2}$, besides, change of δ_1 from 0.01 to 1 did not result in substantial change of the result.

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