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Physical basis of the optical non-destructive method of testing of the gas composition in the cells used in quantum sensors

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We consider phenomena that significantly affect the accuracy of the spectral method of non-destructive optical quality control of cells with alkali metal vapor and buffer gases used as sensitive elements of miniature quantum sensors - frequency standards, magnetometers, gyroscopes. The method makes it possible to determine the composition of a one- or two-component gas mixture based on the results of measuring the broadening and shift of the optical spectral lines of an alkali metal by collisions with a buffer gas. In this work, using ^{87}Rb as an example, we demonstrate that the results of such measurements are affected by distortions of line contours resulting from optical hyperfine pumping. A theoretical description of these effects and ways to eliminate the limitations they introduce are proposed.

Keywords: Spectroscopy of alkali metals, optical control of gas composition, optical pumping, balance equations for populations.

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Development of compact sources of laser pumping, namely, semiconductor lasers with distributed feedback, and then — external-cavity lasers and vertical-cavity surface-emitting-lasers [1] enabled development of miniature optical quantum sensors, such as micro-sized frequency standards [2], quantum magnetometers [3] and gyroscopes [4,5]. The main sensitive element in all these devices is a transparent cell, containing atoms of alkali metal (AM) and a buffer inert gas or a mixture of inert gases. Mass production of miniature frequency standards assigned the industry the task to produce gas cells with strictly maintained parameters — such as total pressure of buffer gas and partial pressure of the components of gas mixture [6,7]. It was found that the smaller is the cell size, the more complicated it is to provide for the required accuracy of its filling, and the errors are greater. A need arose for the non-destructive testing method, which in a hermetically sealed cell may only be conducted by spectroscopic means [8,9]. Since every gas causes AM spectral absorption line shift and broadening that are unique to it, the study of these parameters in case of one- or two-component gas mixture may in principle provide the complete information on its composition [10].

Such testing method includes scanning of narrow-band laser light in the vicinity of AM spectral lines and detection of absorption in the cell. Typical values of shift and broadening coefficients do not exceed 10^7 Hz/Torr [9], and the difference of the shift and broadening coefficients for gases included into the mixture composition may be below 10^6 Hz/Torr, therefore, for the required precision 1 Torr the widths and central frequencies of the absorption lines must be determined with precision ~ 1 MHz at typical line widths of the line 400 MHz and more. This determines the requirements to the signal-to-noise ratio in process of measurement and, accordingly, forces an increase in the light intensity. In this case, at shown below, the parameters of the absorption line turn out to be distorted by the optical pumping processes. The ground state of the AM atom is split into two hyperfine levels, which are characterized by a long relaxation time in the gas cell [11,12]. Therefore, for bleaching the medium due to hyperfine optical pumping, an intensity is required that is many orders of magnitude lower than for bleaching due to saturation of the optical transition. The value of bleaching turns out to be different along the the spectral line profile, which causes its distortion.

This paper devoted to the study of these effects and development of the methods for their calculation and compensation. The theoretical part consisted in determination of the absorption coefficient by solving a system of equations that describe the populations of Zeeman sublevels in the ground state of rubidium in presence of the optical pumping [13].

Let us consider (fig. 1, *a*) the scheme of sublevels in the ground state of AM — rubidium ^{87}Rb (solution for Cs, K etc. is found in a similar way). Every hyperfine level $F = I \pm 1/2$ (where I — nuclear spin, for ^{87}Rb $I = 3/2$) ground $S_{1/2}$- and excited $P_{1/2}$-states in magnetic field is split into $2F + 1$ sublevels with projections of moment $m_F = -F \dots F$. The total quantity of these sublevels is equal to $N = 2(2I + 1)$ (in case of Rb $N = 8$). At temperatures near to room temperature they are in thermal equilibrium, which may be disturbed by optical pumping. In cells with gas mixes already at pressures of around

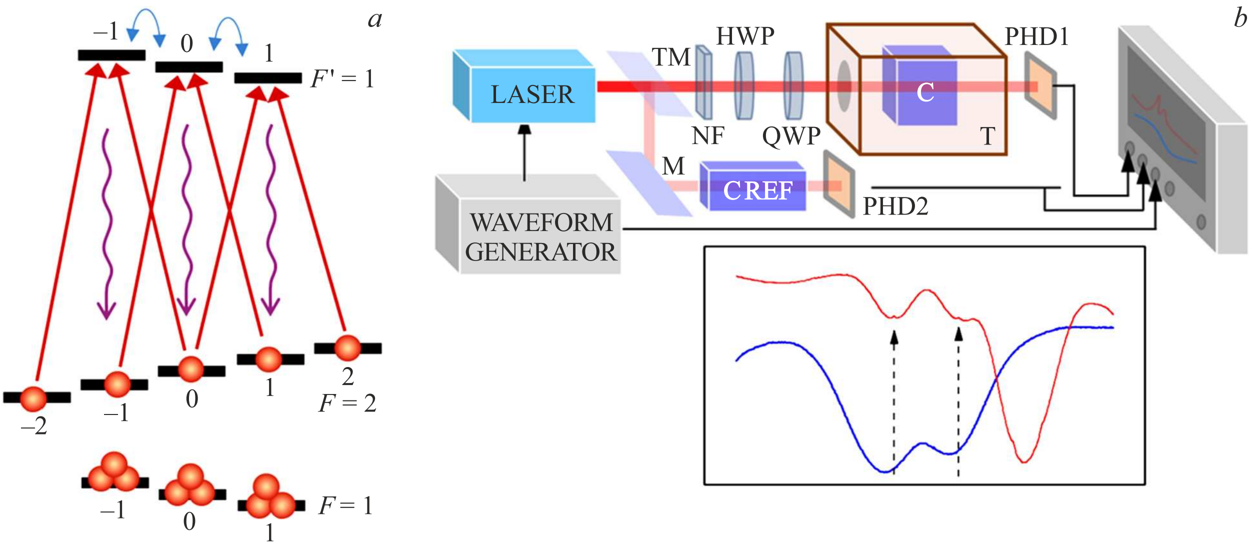

Figure 1. (a) Scheme of Rb levels under conditions of optical pumping by light of circular polarization ($F = 2 \to F' = 1$) and mixing of sublevels of excited state; straight arrows — pumping, wavy ones — relaxation; (b) experimental setup: LASER — diode laser with an external resonant cavity, C — cell, T — thermostat, CREF — reference cell, NF — neutral filter, HWP — half-wave plate, QWP — quarter-wave plate, TM — semi-transparent mirror, M — mirror, PHD1,2 — photodiodes. Inset — an example of passed light intensity recording in the studied cell (thick blue line) and in the reference cell (red line). Dashed arrows show resonances of saturated absorption in the reference cell, corresponding to the central frequencies of transitions $F = 2 \to F' = 1, 2$.

30 Torr the so called mixing of excited state sublevels occurs; as a result, the speeds of of population of the ground state sublevels turn out to be the same, and the difference of their populations is only due to the difference in their depopulation speeds (so-called „depopulation pumping") [14]. In this case, the excited state may be treated as one level with population n_e and decomposition speed Γ. As a result we obtain a system consisting of $N + 1$ linear balance equations

$$\begin{cases} \frac{d}{dt}n_i = -K^i(\nu)In_i - \gamma n_i + \frac{\gamma}{N}\sum_{i=1}^{N} n_i + \frac{\Gamma}{N}n_e = 0, \\ \frac{d}{dt}n_e = -\Gamma n_e + \sum_{i=1}^{N} K^i(\nu)In_i = 0, \end{cases} \quad (1)$$

where $i = 1, \ldots, N$, n_i — levels populations, γ — speed of "dark" relaxation, $K^i(\nu)I$ — speed of optical depopulation pumping from level i. The normalization condition $\sum_i n_i + n_e = 1$ is met. Each inert gas causes a shift and broadening of the spectral line that is characteristic only of it and is proportional to the pressure. Therefore, the composition of the buffer gas in the cell is determined by selection of the distribution type, which affects the speed of optical depopulation pumping $K^i(\nu)I$, and its parameters. Next, the expressions were derived, which describe the shape of the absorption line under slow scanning of the optical pumping frequency, and it is shown that the effects of hyperfine and Zeeman pumping cause non-linear change of absorption and, accordingly — the effective broadening and shift of spectral lines. It should be especially noted that the mentioned broadening is not related to light power broadening and is implemented at intensities that are many times smaller than the intensity of optical saturation.

To test the theoretical results, the experimental research was done on dependences of absorption line parameters in the cell on the parameters of the detecting light. For this purpose the studied cell that contained saturated vapors ^{87}Rb and gas mix, were placed in a thermostat (fig. 1, b), and radiation of a laser with an external resonant cavity (VitaWave, Moscow) was scanned in the vicinity of the transitions that form D_1-line of absorption for Rb (795 nm).

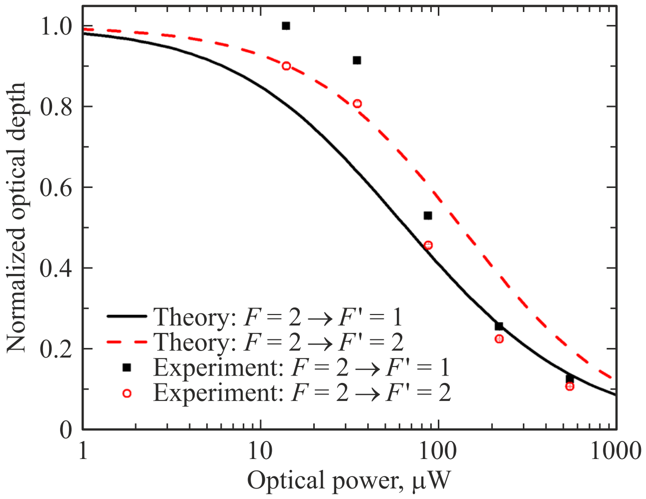

Figure 2. Result of theoretical estimation (lines) and experimental measurement (symbols) of optical density dependence in the center of spectral lines $F = 2 \to F' = 1, 2$ ^{87}Rb on the power of the detecting light.

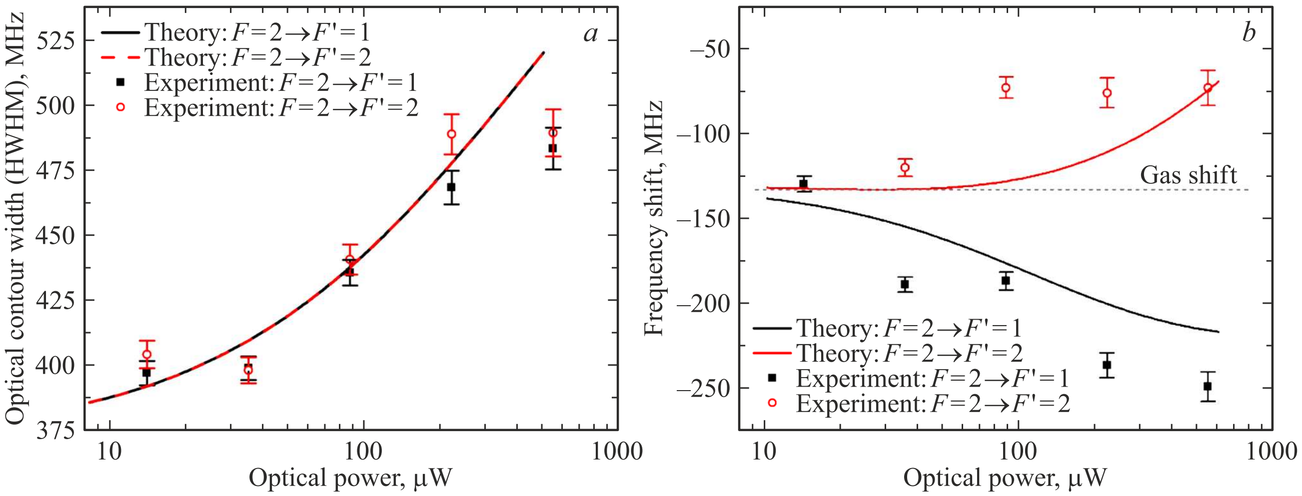

Figure 3. Dependences (*a*) of effective broadening and (*b*) shift of spectral lines $F = 2 \to F' = 1, 2$ ^{87}Rb on the power of detecting radiation: lines — theoretical estimate, symbols — experiment.

Simultaneously, in the Rb reference vacuum cell resonances of non-linear saturated absorption were recorded [15]. Their width was ~ 5 MHz, which, when the signal exceeded noise by an order, made it possible to determine the laser frequency with the accuracy of around 0.5 MHz. When the spectra were processed, the basic line was subtracted, and the contours were approximated with the sum of Gaussian profiles, which under these conditions described well the central part of the absorption lines. This approximation is acceptable as long as the broadening by buffer gas is comparable in value with Doppler broadening, but for high gas pressures, it is necessary to use a Lorentzian or a Voigt contours. The results of comparing the dependences of the effective broadening and shift and the optical density in the center of the spectral line of the detecting light power are given in fig. 2, 3.

Here it should be explained that the cell chosen for a demonstration experiment should have met several criteria: 1) it should have contained one AM isotope, 2) buffer gas pressure therein should be rather high to provide for mixing of the excited state levels, 3) collisional broadening therein should have been substantially lower than hyperfine splitting of the excited state levels (814 MHz). It is easy to see that criteria 2 and 3 are hard to be met together; besides, at pressure meeting such criteria, the collisional broadening becomes close to the Doppler one, and approximation of the absorption contours must be described by the Voigt contour. At this stage of the paper we limited ourselves to Lorentz and Gaussian contours, and used a model of full mixing of levels, which explains minor differences in the theory and the experiment. Note that the area of practical application of the method is much wider that the area defined by criteria 1–3.

From fig. 3 it follows that hyperfine pumping causes effective broadening and shift of optical spectral lines, besides, the broadening value substantially exceeds the direct broadening of the transition with optical power (so called light power broadening). Demonstration of impact of the hyperfine pumping at contours of AM spectral lines is an indirect evidence that this impact may be eliminated the „re-pumping" method (i.e. depopulation pumping from the second hyperfine level), widely used in laser cooling to increase its intensity [16]. Besides, polarization of the detecting light must be selected to minimize the impact of Zeeman pumping, for example, linear polarization perpendicular to magnetic field (fig. 1, *a*).

Thus, we made the theoretical estimate and tested it by comparison with the experiment regarding the distortions of the rubidium spectral line contours in presence of the buffer gas caused by optical hyperfine pumping in the approximation of full mixing of Zeeman levels populations in the excited state. It is shown that the parameters of the absorption line when measured turn out to be distorted by the processes of optical pumping, causing effective broadening of the spectrum of optical transitions, and their shift as a result of the presence of closely located adjacent lines in the Rb and Cs spectra. The obtained results indicate the possibility of correcting the results of direct measurement with theory data, or (in the future) the possibility to eliminate such distortions by the repumping method (hyperfine depopulation pumping. These results also open the possibility to use the optical spectroscopic method for non-destructive testing of the gas composition of miniature and microminiature cells.

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Conflict of interest

The authors declare that they have no conflict of interest.

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