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Sub-Doppler resonances caused by photoionization of atoms in thin gas cells

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By analogy with the well-tested method of high-resolution spectroscopy in thin gas cells for the processes of optical pumping of atoms, this work shows the possibility of detecting narrow sub-Doppler optical resonances caused directly by straight photoionization of atoms (or molecules) in such cells. The structure of the established nontrivial resonances substantially depends on the probability of photoionization of atoms and dimensions of such a cell, the internal thickness of which is many times smaller than its diameter. Of particular interest is the broadening of the considered sub-Doppler resonances, which is determined directly by the photoionization cross section of atoms and the intensity of radiation causing ionization. Under certain conditions, such photoionization broadening can be measured experimentally with high accuracy, as a result of which it is possible to obtain new important information about ionization processes in atoms and molecules.

Keywords: sub-Doppler resonances, photoionization of atoms, thin gas cell, transit-time relaxation of atoms.

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Introduction

Research of photoionization processes in rarefied gas media is critical for atomic and molecular spectroscopy, plasma physics and astrophysics [1–4]. Direct single-photon ionization from the ground atomic (molecular) quantum state may be realized by ultraviolet or vacuum ultraviolet radiation, which, in particular, is generated by synchrotrons and some gas discharge lamps, and is also present in solar radiation [1–3]. At the same time multi-photon ionization of atoms is done by rather intense laser radiation of visible or infrared ranges [4].

Effectiveness of direct photoionization of individual atoms (or molecules) is determined both by intensity of ionizing radiation and time of its action on a particle. Therefore, if the gas medium in a cell is rarefied enough, the photoionization process will mostly depend on specific transit times of atoms (molecules) between the walls of such a cell. Similar situation is specific for optical pumping of atoms, when light-induced redistribution of their ground quantum term sublevel populations takes place [5]. The papers [6–9] proposed new methods of sub-Doppler laser spectroscopy based on specific nature of stationary optical pumping of atoms during their transit between walls of a thin gas cell, whose specific transverse size D is many times more than its internal thickness L. Further these methods were successfully realized on experiments for precision spectral analysis of atoms [10–15] based on a series of thin cells (with different internal thickness up to $10\,\mu$m), containing vapors of cesium [10–14] or rubidium [15]. Paper [16] presents the review of such methods, achievements and possibilities of ultrahigh resolution spectroscopy in such cells. In particular, the method of sub-Doppler spectroscopy was developed and tested, where optical pumping of atoms may be implemented even by broad-band emission in the entire volume of the thin cell, and their probing is done with a probe monochromatic light beam [7,9,12–16].

This paper shows that such spectroscopy method may be also used to analyze photoionization of atoms (molecules) of rarefied gas. Indeed, let us consider the experiment scheme in fig. 1, where direct photoionization of atoms is carried out in the entire volume of the thin cell from their ground quantum state a to continuum state i (fig. 2). In the considered situation of rather rarefied gas the reduction in quantity of atoms at the level of a due to ionization process is determined by time of their transit between cells walls, where atom particles relax to equilibrium state. In our case such relaxation is mainly caused by collisions of atoms with end (plane-parallel) walls of the thin cell (fig. 1). Therefore, processes of direct photoionization and transit relaxation of atoms in such cell result in a narrow dip in the distribution of electrically neural atoms by projection v_z of their velocity along axis z of the cell to the surroundings of value $v_z = 0$. Therefore, sub-Doppler resonances may arise in the spectrum of monochromatic light wave absorption with wave vector $\mathbf{k}$, directed along axis z (fig. 1), at the central frequencies of optical transitions from the ground quantum state a to any excited levels of electrically neutral atoms (fig. 2). This paper shows that the structure of such detected narrow spectral resonances will depend considerably on the probability of the photoionization process. As a result, new opportunities open to determine the cross section of photoionization of atoms (molecules) and intensity

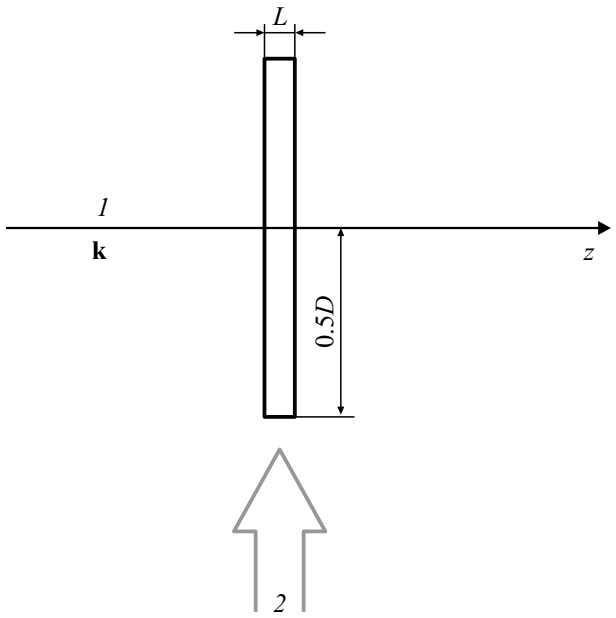

Figure 1. Scheme of experiment with thin gas cell with diameter D and inner thickness $L \ll D$, where *1* is the probe monochromatic light beam, *2* is the ionizing radiation.

of ionizing radiation by ultrahigh resolution spectroscopy methods.

Basic relations

Let us consider direct photoionization of atoms (or molecules) of gas medium with stationary radiation, homogeneous in the entire volume of the thin cylindrical gas cell (fig. 1), which is made of material transparent both for ionizing and probe radiations. It is suggested that in this rather rarefied gas layer one may neglect the interatomic interaction. Weak probe monochromatic light wave with wave vector $\mathbf{k}$ and comparatively small diameter $d \ll D$ propagates through cell center along axis z (fig. 1). Frequency ω of this wave is scanned in the surroundings of center ω_0 of a certain optical transition $a \to b$ from the ground quantum level a to excited level b of electrically neutral atoms (fig. 2). Under considered conditions, the following balance equation is present for population $\rho_a(\mathbf{r}, \mathbf{v})$ of atoms in the ground state a with speed $\mathbf{v}$ and coordinate vector $\mathbf{r}$:

$$\mathbf{v}\frac{\partial \rho_a}{\partial \mathbf{r}} = -\xi \rho_a, \qquad (1)$$

where ξ is the probability (per unit of time) of direct photoionization of atoms from state a (fig. 2), which is determined by intensity of ionizing radiation and cross section of photoionization. Equation (1) must be supplemented with boundary conditions depending on the features of collisions of electrically neutral atoms and produced photoions with walls of the cell. Same as in calculations [6–16] made for the processes of optical pumping in thin gas cells, we will believe that such collisions lead to equilibrium distribution both by velocities of atoms and by populations of their quantum levels. It is important to note that the intensity of ionization considered is not too high, when the number of arising photoions and photoelectrons is many times lower than the number of atoms in the cell. As a result of the ion recharging process on the surface [17] we shall assume that after several collisions of a photoion with cell walls it is neutralized, i. e. the atom is restored. We also neglect the influence of the arising photoelectrons to distribution of electrically neutral atoms of the gas layer. Then using equation (1) we obtain the following formula for population $\rho_a(\mathbf{r}, \mathbf{v})$ of the ground level a of atoms in the region of the probe light beam concentrated around the central axis of the cell z (fig. 1) in the interval of $-0.5L \le z \le 0.5L$:

$$\begin{aligned}\rho_a(z, v_z, v_r) &= n_a F_l(v_z) F_r(v_r) \\ &\times \left\{ \left[\exp\left(-\xi \frac{(z+0.5L)}{v_z} \right) \eta\left(\frac{Dv_z}{2v_r} - z - 0.5L \right) \right.\right. \\ &+ \left. \exp\left(-\xi \frac{D}{2v_r} \right) \eta\left(z + 0.5L - \frac{Dv_z}{2v_r} \right) \right] \eta(v_z) \\ &+ \left[\exp\left(\frac{-\xi(z-0.5L)}{v_z} \right) \eta\left(z - 0.5L - \frac{Dv_z}{2v_r} \right) \right. \\ &+ \left.\left. \exp\left(-\xi \frac{D}{2v_r} \right) \eta\left(\frac{Dv_z}{2v_r} - z + 0.5L \right) \right] \eta(-v_z) \right\}, \qquad (2)\end{aligned}$$

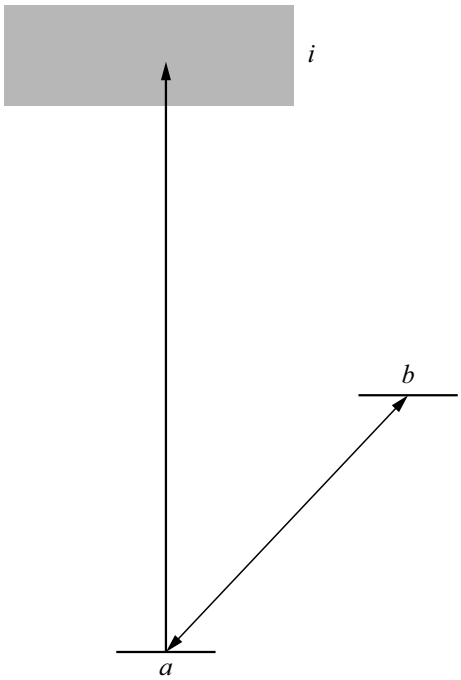

Figure 2. Scheme of considered quantum levels and transitions: $a \to i$ is the direct photoionization of atoms from the ground term a to state of ionizing continuum i, $a \to b$ is the optical transition from state a to excited state b, probed by the monochromatic light beam.

where $\eta(x)$ is the stepwise function ($\eta(x) = 1$, if $x \geq 0$ and $\eta(x) = 0$, when $x < 0$), n_a is the equilibrium density of atoms at the level of a with Maxwell distributions $F_l(v_z)$ and $F_r(v_r)$ accordingly along longitudinal v_z and radial v_r components of atom velocity $\mathbf{v}$:

$$F_l(v_z) = \pi^{-0.5} u^{-1} \exp(-v_z^2 u^{-2}),$$

$$F_r(v_r) = 2 v_r u^{-2} \exp(-v_r^2 u^{-2}), \qquad (3)$$

with the most probable speed u of atoms in gas. From equation (2) we obtain the following distribution by projection of velocity v_z for averaged population $N_a(v_z)$ of atoms concentrated around the central axis of the cell z (fig. 1):

$$N_a(v_z) = L^{-1} \int_{-0.5L}^{0.5L} \left[\int_0^{\infty} \rho_a(z, v_z, v_r) dv_r \right] dz. \qquad (4)$$

Further, as in papers [6–16] with stationary optical pumping, we will assume that cell thickness $L \gg 1\,\mu$m, i. e. many times exceeds the optical radiation wave length. Then the linear coefficient of absorption G of weak probe optical wave at transition $a \to b$ (fig. 2) in the considered gas cell has the form of

$$G(\delta) = G_0 \frac{\{\int_{-\infty}^{\infty} N_a(v_z)[\gamma^2 + (\delta - k v_z)^2]^{-1} dv_z\}}{\{\int_{-\infty}^{\infty} n_a F_l(v_z)[\gamma^2 + (k v_z)^2]^{-1} dv_z\}}, \qquad (5)$$

where population $N_a(v_z)$ is determined by formula (4), $k = |\mathbf{k}| = \omega/c$, $\delta = (\omega - \omega_0)$ is the detuning of scanned frequency ω from center ω_0 of optical transition $a \to b$ (fig. 2) with natural half-width γ and Doppler widening $ku \gg \gamma$ of spectral line, multiplier G_0 is the coefficient of absorption of the probe wave at this transition $a \to b$ in absence of ionizing radiation and at zero detuning $\delta = 0$.

Discussion of results

Fig. 3, *a* presents distribution $N_a(v_z)$ (4) of atom population at the ground level a on atomic projection of velocity v_z at various probabilities of photoionization ξ and ratios $D/L \gg 1$ of thin cell dimensions (fig. 1). It is seen that in the dependence $N_a(v_z)$ on the background of quite broad velocity distribution with specific width determined by most probable atom speed u, there is a narrow symmetric dip with the center in point $v_z = 0$. This dip is caused by specific nature of direct ionization and transit relaxation of atoms in the considered cell. Its origin is well-understood in a model of a plane one-dimensional cell with size ratio $D/L \to \infty$. Then, according to equation (1), effective ionization of atoms is carried out under condition

$$\xi \frac{L}{|v_z|} \geq 1, \qquad (6)$$

i. e. at rather long transit time $L/|v_z|$ of atoms between plane parallel walls of the cell. From (6) it directly follows that atom ionization process sharply increases at $|v_z| \to 0$, as a result of which a narrow central dip occurs in distribution $N_a(v_z)$ in fig. 3, *a*.

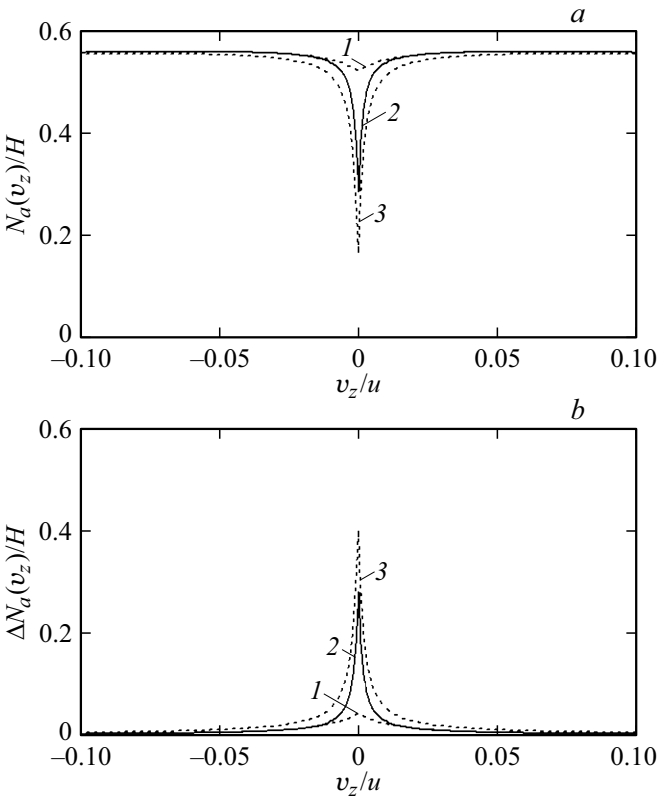

Figure 3. Distributions $N_a(v_z)$ (*a*) and $\Delta N_a(v_z)$ (*b*) of atoms in the ground quantum state a by component of atom velocity v_z (in units u), when $\xi(L/u) = 0.001$ (*1, 2*) and 0.002 (*3*), and $(D/L) = 100$ (*1*) and 1000 (*2, 3*). These functions $N_a(v_z)$ and $\Delta N_a(v_z)$ are normalized by value $H = n_a u^{-1}$.

In case of a gas cell with real transverse dimension D for effective photoionization of atoms together with ratio (6) the following condition shall be met for their radial component of velocity v_r:

$$\xi \frac{D}{2|v_r|} \geq 1. \qquad (7)$$

Such condition is related to specific time $0.5D/|v_r|$ of atom transit in transverse direction to the central area of the cell probed by the probe light beam (fig. 1).

According to ratios (6), (7), the considered central dip in distribution $N_a(v_z)$ in fig. 3, *a* increases with growing probability of photoionization ξ and diameter D of thin cell (with its fixed inner thickness L).

The number of photoionized atoms in the cell is determined by value $\Delta N_a(v_z)$:

$$\Delta N_a(v_z) = n_a F_l(v_z) - N_a(v_z). \qquad (8)$$

Corresponding dependencies $\Delta N_a(v_z)$ are presented in fig. 3, *b* and directly reflect the structure of the dip in the distribution $N_a(v_z)$ in fig. 3, *a*. Notice that integral quantity

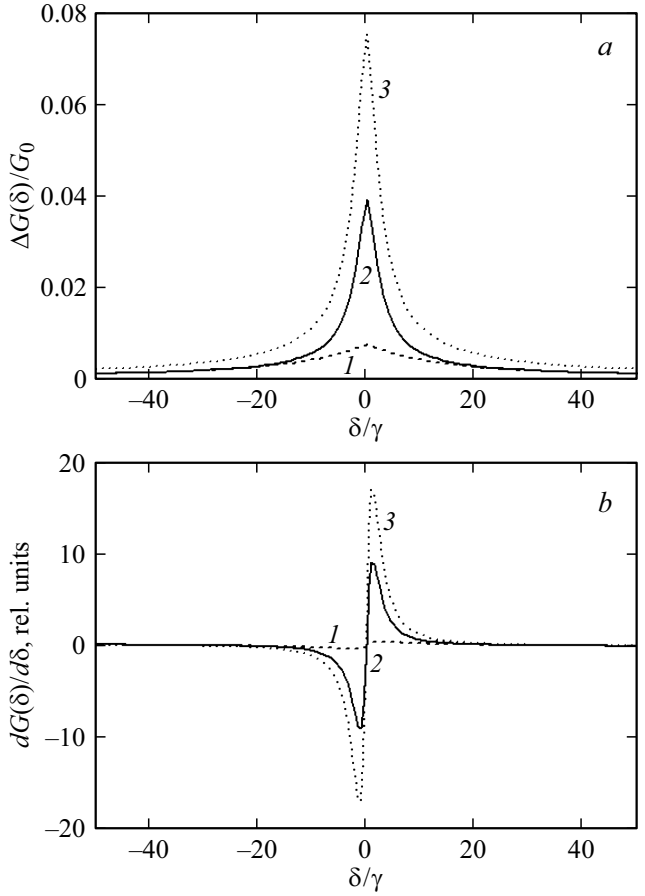

Figure 4. Sub-Doppler resonances $\Delta G(\delta)$ (a) and $\frac{dG(\delta)}{d\delta}$ (b) depending on detuning of frequency δ (in units γ), when $\gamma = 10^{-3}ku$, $\xi(L/u) = 0.0001$ $(1, 2)$ and 0.0002 (3), and $(D/L) = 100$ (1) and 1000 $(2, 3)$.

$\int_{-\infty}^{\infty} \Delta N_a(v_z)dv_z$ of produced photoions and photoelectrons in our case is many times less than the quantity of electrically neutral atoms.

A narrow central dip in velocity distribution $N_a(v_z)$ (fig. 3, a) results in sub Doppler resonance with the center at zero frequency detuning $\delta = 0$ in the spectral dependence of absorption ratio $G(\delta)$ (5) of the probe monochromatic light beam. Such narrow resonances may, in particular, be observed as difference between signals of the probe wave absorption that are sequentially recorded in presence and in absence of ionizing radiation (fig. 1). Then the detected part $\Delta G(\delta)$ of absorption ratio is produced from formula $G(\delta)$ (5) by substitution of $N_a(v_z)$ with $\Delta N_a(v_z)$ (8). Another method to record such sub-Doppler resonances is possible by quite low modulation of probe radiation frequency, when the frequency derivative $dG(\delta)/d\delta$ of absorption ratio $G(\delta)$ (5) is directly identified. Such spectral dependencies $\Delta G(\delta)$ and $dG(\delta)/d\delta$ are shown in fig. 4. Same as the corresponding central dip in speed distribution $N_a(v_z)$ (fig. 3), the considered sub-Doppler structures in fig. 4 increase with growing probability of photoionization ξ and ratio D/L of thin cell dimensions (fig. 1). Analysis of specific widths of such sub-Doppler resonances is of special interest, as they may be measured in an experiment with rather high accuracy by known methods of optical spectroscopy [5]. In case of resonance $\Delta G(\delta)$ (fig. 4, a) with amplitude $A_1 = \Delta G(\delta = 0)$ let us consider such width w_1 as its value at half-height of dependence $\Delta G(\delta)$. For sub-Doppler resonance of dispersion form in fig. 4, b we will define its specific width w_2 by frequency interval between maximum and minimum of function $dG(\delta)/d\delta$, and its amplitude A_2 — by difference between such extrema.

Fig. 5, a and 6, a present dependencies of widths w_1 and w_2 of investigated sub-Doppler resonances (fig. 4) on probability ξ of atom photoionization at various ratios D/L of thin cell dimensions with fixed inner thickness L (fig. 1). Curves *3* in fig. 5 and 6 correspond to the model of plane one-dimensional cell, when $D/L \to \infty$. At extremely low probability of photoionization $\xi(L/u) \to 0$

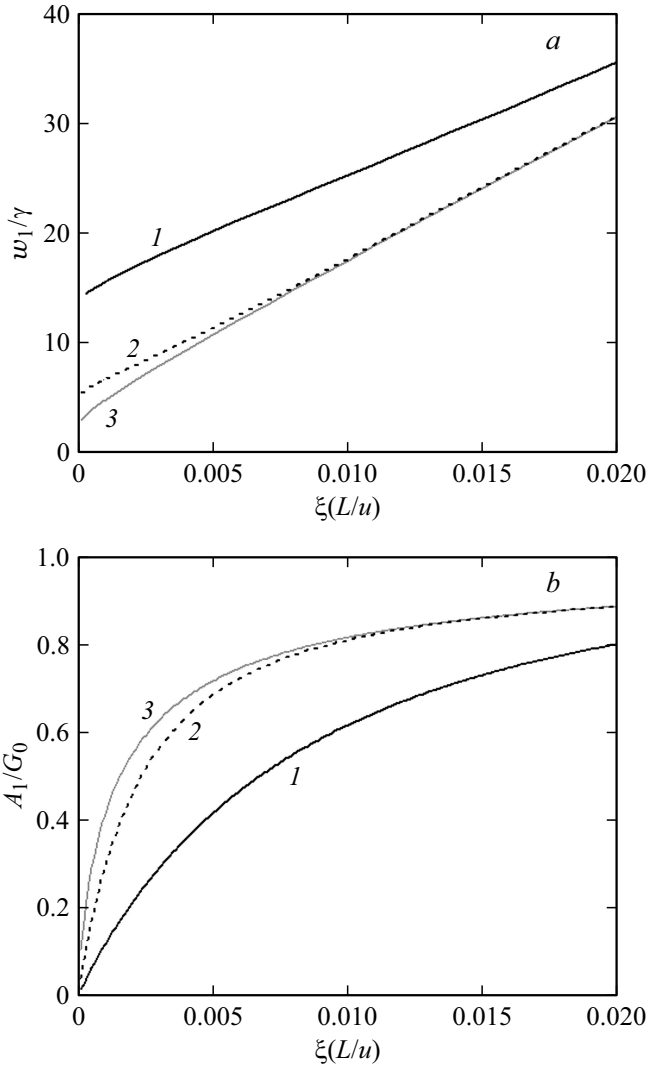

Figure 5. Dependencies of width w_1 (a) and amplitude A_1 (b) of sub-Doppler resonance $\Delta G(\delta)$ on probability of photoionization ξ (in units u/L), when $\gamma = 10^{-3}ku$, $(D/L) = 200$ (1), 1000 (2) and ∞ (3).

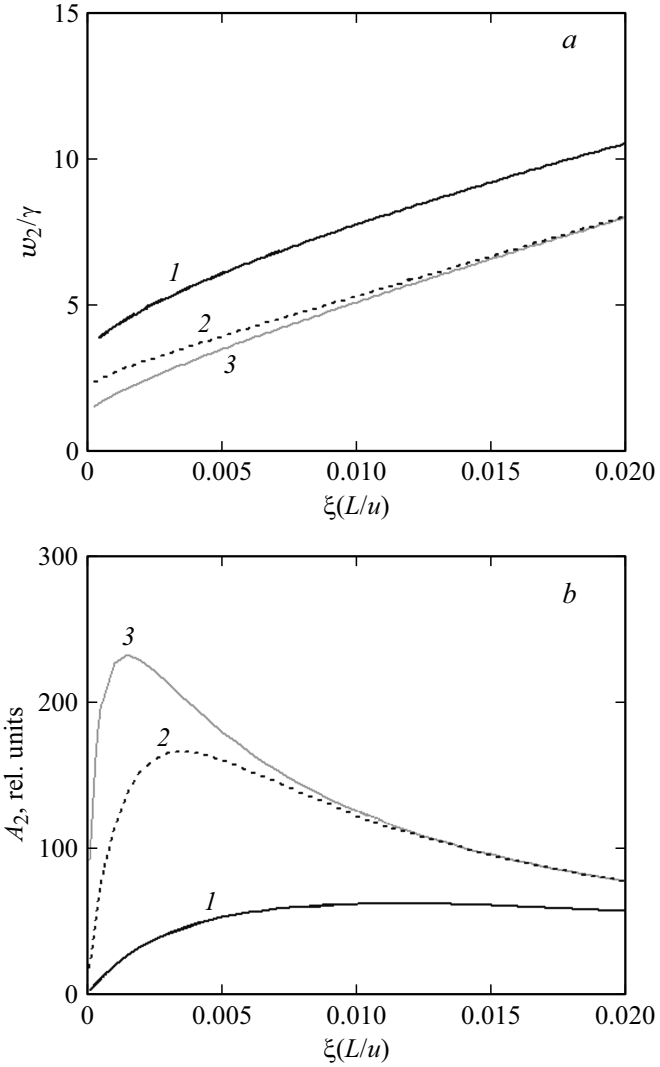

Figure 6. Dependencies of width w_2 (a) and amplitude A_2 (b) of sub-Doppler resonance $dG(\delta)/d\delta$ on probability of photoionization ξ (in units u/L), when $\gamma = 10^{-3}ku$, $(D/L) = 200$ (*1*), 1000 (*2*) and ∞ (*3*).

the widths described by curves *3*, asymptotically approach values $w_{10} \approx 2.5\gamma$ (fig. 5, a) and $w_{20} \approx 1.25\gamma$ (fig. 6, a), which comply with natural broadening of considered sub-Doppler resonances. In case of cells with real transverse dimensions D (fig. 1) such resonances will also demonstrate residual Doppler broadening of the order of $(L/D)ku$ (curves *1* and *2* in fig. 5, a and 6, a). Indeed, according to the velocity distribution $\Delta N_a(v_z)$ in fig. 3, b, the collection of photoionized atoms is a compact analogue of atom beam with specific divergence $\sim (L/D)$. Therefore, as ratio D/L decreases, the corresponding resonance widths increase in fig. 5, a and 6, a. Atom ionization causes additional broadening of such resonances. Starting from certain probability values ξ, such photoionization broadening becomes dominant, as a result the difference between widths of sub-Doppler resonances produced at various ratios of cell dimensions $(D/L) \gg 1$ (curves *2* and *3* in fig. 5, a and 6, a) disappears asymptotically. Then dependencies of widths w_1 and w_2 on the probability of photoionization ξ are of quais-linear nature.

According to fig. 5, b, amplitude A_1 of sub-Doppler resonance $\Delta G(\delta)$ increases with growth of photoionization probability ξ and diameter D of the thin cell (with its fixed inner thickness L). This is compliant with dependence $\Delta N_a(v_z)$ (8), shown in fig. 3, b. At the same time as photoionization probability ξ increases, amplitude A_2 of function $dG(\delta)/d\delta$ first increases to a certain value, and then it decreases monotonously (fig. 6, b). At rather low values ξ dependencies of amplitudes $A_1(\xi)$ and $A_2(\xi)$ are of quasi-linear nature (fig. 5, b and 6, b).

Conclusion

This paper shows possibility to record sub-Doppler resonances caused by direct photoionization of atoms (or molecules) and features of their transit relaxation in thin cells with rarefied gas medium. Such recording may be carried out similarly to the ultrahigh resolution spectroscopy method, developed and tested previously for the processes of optical pumping of atoms in such cells [7,9,12–16]. The structure of considered sub-Doppler resonances considerably depends on the probability ξ of atom photoionization per unit of time, and also on ratio $(D/L) \gg 1$ of thin gas cell dimensions (fig. 1). Photoionization broadening of these resonances is of special interest, as it increases with growth of value ξ and may be measured in an experiment with high accuracy using known methods of optical spectroscopy. For this purpose it is necessary that such photoionization broadening is substantial compared to natural and residual Doppler broadening of studied resonances. Such condition is possible, when optical transition $a \to b$ (fig. 2) is selected for the probe monochromatic wave with rather small ratio $\gamma/(ku) \ll 1$ of homogeneous half-width of spectral line γ to its Doppler widening ku. Besides, it is necessary to use thin gas cells with rather small ratio of dimensions $(L/D) \ll 1$ (fig. 1). For example, ratio $(L/D) = 10^{-3}$ set in calculations of fig. 3–6 includes a space for cells with thickness $L \sim 10\,\mu$m for their transverse size $D \sim 1$ cm. Such thin cells with alkaline metal vapors have been used in experiments for many years [10,11]. The method of sub-Doppler spectroscopy proposed in this paper will help to obtain new important information about direct photoionization of atoms and molecules that will supplement the results of already known ionization research methods [1–5].

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