Pressure shift coefficient measurements in an RF discharge for Ar $4s[3/2]$$_2$—$5p[3/2]$$_3$ transition with the help of diode-laser absorption spectroscopy

A K Chernyshov$^{1,2}$, P A Mikheyev$^{1,2}$, N N Lunev$^1$, and V N Azyazov$^{1,2}$

$^1$Lebedev Physical Institute, 443011, Samara, Russia
$^2$Samara National Research University, 443086, Samara, Russia
e-mail: chak@fian.smr.ru

Abstract. Optically pumped all-rare-gas laser (OPRGL) with unique properties were recently proposed with a possibility to obtain the laser power on the order of hundreds of Watts from a cubic centimeter. To provide high laser efficiency, the pumping radiation has to match the absorption spectrum of the rare gas metastables. To meet this condition a reliable diagnostics of the key parameters of the active medium is required and knowledge of the broadening and shift coefficients for corresponding transitions of rare gases is necessary. In this paper, the diode-laser absorption spectroscopy was employed to determine the pressure shift coefficient for 811.5 nm Ar line. The value of obtained coefficient in pure argon reduced to 300 K is $-(2.1 \pm 0.1) \times 10^{-10}$ s$^{-1}$cm$^3$. In the course of the study the pressure broadening coefficient was also evaluated and found to be $(2.4 \pm 0.5) \times 10^{-10}$ s$^{-1}$cm$^3$.

Keywords: Optically pumped all-rare-gas laser, metastable argon atoms, radio-frequency discharge, diode-laser absorption spectroscopy, spectral line pressure shift

1. Introduction

The well-known diode-pumped alkaline-vapour lasers (DPAL) attracted considerable attention because of their possibility to obtain optical power at a level of several kilowatts from a volume of a few tens of cubic centimetres [1]. At the same time, these lasers possess intrinsic technical difficulties associated with high reactivity of the alkali metals. As a chemically passive alternative to DPAL, in 2012 lasing was demonstrated employing metastables of the heavier rare gases (neon argon, krypton and xenon) [2, 3]. In such systems, both electric discharge and radiation from high-power diode lasers are supposed to be used. In this case, the electric discharge provides the population of the lower (metastable) laser level, the optical pumping populates the upper pump level and subsequent collisional energy transfer populates the upper laser level. These lasers are now called optically pumped rare gas lasers (OPRGL), they operate according to the traditional three-level scheme.

A high lasing threshold characterizes the three-level laser scheme. Model estimates of an OPRGL show that to obtain a specific optical power of the order of 100 W/cm$^3$, it is necessary that the number density of metastable atoms to be on an order of $10^{12}-10^{13}$ cm$^3$ at a gas mixture pressure of about 1 atm [4]. In addition, the radiation of diode lasers providing optical pumping must be spectrally

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd
matched with the absorption line of the rare gas metastables. To meet these conditions the reliable diagnostics of the laser active medium is needed. It means, first of all, that the key parameters such as metastables’ number density and the pressure broadening/shift coefficients for the transitions of interest are known.

Earlier, we measured the pressure-broadening coefficients of absorption lines of argon and krypton atoms with the help of diode-laser absorption spectroscopy [5-7]. In our experiments, the metastable atoms were generated in plasma of an RF discharge for gas mixtures that are commonly used for OPRGL. In this paper, the previously used spectroscopic setup was upgraded, which allowed us to measure the pressure broadening and the shift coefficients of absorption lines in rare gases simultaneously. The above mentioned coefficients for the $4s[3/2]_2 \rightarrow 5p[3/2]_3$ transition of the Ar atom were measured upon excitation by a 40 MHz RF discharge in pure argon.

2. Description of the method
Dependence of the shift of an absorption line $\Delta\nu_{sh}$ on pressure $P$ (Torr) and temperature $T$(K) is as follows [8]

$$\Delta\nu_{sh}(P,T) = 3.22 \cdot 10^{16} \gamma(T_0) \left(\frac{T_0}{T}\right)^{0.7} P.$$  (1)

Here $T_0$ is the reference temperature chosen equal to 300 K. As follows from (1), to determine the shift coefficient $\gamma(T_0)$ it is necessary to measure three quantities, namely the shift of the spectral line $\Delta\nu_{sh}$, gas temperature $T$ and pressure $P$ in a gas discharge cell. It is easy to measure gas pressure $P$ by means of the commercially available device and it is the most reliably determined quantity from the specified above parameters set.

In usual spectroscopic examinations gas temperature $T$ in a sample cell is equal to temperature of the ambient air and changes in time only slightly, therefore this parameter does not create any problems during measurements. On the other hand, in plasma spectroscopy, gas temperature in a discharge noticeably differs from ambient air temperature and can vary considerably owing to changes of excitation conditions in an RF discharge. For example, gas temperature $T$ in a discharge can vary considerably with changes in pressure $P$ and that it is necessary to consider.

The temperature of a neutral gas $T$ in plasma is convenient to determine by non-invasive probing from a profile of the spectral line. For this purpose, the measured profile of the absorption line was fitted numerically by the Voigt function, which is a convolution of the Doppler and Lorentz contours. Numerical fitting to the Voigt function allowed to reconstruct the widths (FWHM) of the Doppler $WD(T)$ and the Lorentz $WL(P, T)$ contours simultaneously in accordance with the technique [6, 7]. In turn, the width $WD(T)$ of the Doppler contour is uniquely determined by the gas temperature $T$ in accordance to the known expression [9]

$$WD(T) = 7.16 \cdot 10^{-7} v_0 \left(\frac{T}{M_r}\right)^{1/2},$$  (2)

where $M_r$ is the mass of the Ar atom in atomic units. Then for the reference temperature $T_0$, the width of the Doppler contour of argon will be equal to $WD(T_0) = 0.724$ GHz. Using equation (2), expression (1) for the line shift $\Delta\nu_{sh}$ can be rewritten in another form

$$\Delta\nu_{sh}(P,T) \left(\frac{WD(T)}{0.724}\right)^{-1.4} = 3.22 \cdot 10^{16} \gamma(T_0) P.$$  (3)

As a result of such a transformation, a combination of experimentally measured quantities on the left-hand side of relation (3), exhibits linear dependence on gas pressure $P$. The slope of the line given by the expression (3) is related to the pressure shift coefficient $\gamma(T_0)$. 


The dependence of the width of the Lorentz contour $WL(P, T)$ on the temperature $T$ and pressure $P$ is similar to that for the line shift $\Delta \nu_{sh}(P, T)$ [6]

$$WL(P, T) \left( \frac{WD(T)}{0.724} \right)^{1.4} = 3.22 \cdot 10^{16} \cdot 2\xi(T_0)P.$$ (4)

Since the coefficient $\xi(T_0)_{Ar}$ for argon was previously measured [5, 6], it is possible to determine the gas pressure $P$ from the value $WL(P, T)$ and compare it with the measured value and thus, to verify the correctness of all intermediate actions associated with the definition of $WD(T)$ and $WL(P, T)$.

3. Experimental setup

Diode-laser spectroscopy was used to measure the shape of the argon line. A simplified diagram of the experimental setup, which consisted of three main parts, is shown in figure 1. First, it had a sample cell to which RF electronic units and a rare gas pumping system were connected. The Ar-sample cell is a quartz discharge tube 50 cm long, 15 mm in diameter with a wall thickness of 1.5 mm. The glass windows were fixed at an angle to optical axis at the ends of the discharge tube. External electrodes were attached at the middle of the sample cell, as shown in the inset of figure 1. The electrodes were U-shaped pieces of thick copper wire with 10 cm sides. A symmetrical 2-wire line was connected to the electrodes, through which the RF power (~10 W) from the commercially available 40 MHz generator reached the discharge cell through a matching circuit (not shown).

![Figure 1. Simplified diagram of the experimental setup (on the left) and the fluorescent lamp starter used as a reference cell (on the right). The cross section of the discharge tube with four wire electrodes is shown near to the sample cell within the grounded chamber.](image)

The experiments were performed in the regime of constant gas pumping. Argon (0.999 purity) from the gas cylinder through the reduction gear and the mass flow controller FM (Bronkhorst) was fed to the inlet socket of the cell. The outlet socket of the sample cell was connected to an oil-free vacuum pump (nXDS10i, Edwards) through a needle choke valve. The gas pressure in the discharge tube was regulated by the needle valve and controlled by a capacitive pressure sensor P (Metran) installed near the outlet.

The second component is the source of probing optical radiation, which is a diode laser (DL) L808PO30 (Thorlabs), equipped with a short external resonator of the original configuration [10, 11]. The current/temperature controller ITC4001 (Thorlabs) was used to power the laser and stabilize its
Figure 2. Observation of 811.5 nm line in argon: a) the initial absorption signals with the tilted baseline after passing the appropriate cells and the transmission resonance signals of the reference interferometer (FSR = 750 MHz) recorded versus time; b) Processed absorption signals with a horizontal baseline, as a function of detuning from the atomic line center.

temperature. A triangular signal from an external generator with a frequency of 40 Hz was added into the injection current of the DL through the controller ITC4001 to obtain continuous tuning of the laser frequency. Liquid crystal cell (LC), operating in the quarter wave plate mode, acted as a polarization isolator [12]. The spectrum monitoring system of the laser radiation included a spectrometer (not shown in figure 1) AvaSpec-3648 (Avantes) and a confocal interferometer (F ≈ 200, FSR = 750 GHz). With their help, it was found that the range of the discrete temperature tuning of the laser wavelength was ∼3nm, the continuous current tuning (near 810 nm) of the laser frequency was on the order of 36 GHz. The spectral width of the probing radiation was estimated to be 50 MHz. The maximum optical output power available from the laser diode was up to 5 mW, which was measured with the help of a photodiode power meter (PD300, Ophir).

The third component of the experimental setup was a small sealed-off discharge cell, which was used as a reference for the optical frequency, relative to which the shift of the argon absorption line in the sample cell was determined. It is rather difficult to manufacture a high-quality sealed gas-discharge cell in an ordinary optical laboratory [13]. This is due to the fact that the accumulated concentration of metastables of the rare gas is very sensitive to the presence of contaminants. Therefore, the Ar starter from the fluorescent lamp (S2 220/240V, 4-22W, Philips) was used as the reference cell [14]. To excite the radio-frequency discharge in the glass bulb of the starter, instead of the built-in electrodes, an external ring electrode was used, which simultaneously served as the holder figure 1 (on the right). This ring electrode was connected directly to a low-power 50 MHz transistor generator. The role of the second electrode was performed by the grounded metal case of the device. A thin probing laser beam passed through the starter bulb between the internal electrodes and was registered with a photodetector. Silicon monolithic photodiodes with on-chip transimpedance amplifiers (OPT101, Texas Instruments) were used as photodetectors in the setup.

4. Results and discussion
Figure 2(a) shows the initial signals recorded in the experiment. The sloped line 1 is the base line of zero absorption, recorded by the photodetector PD1 which was installed at the entrance to the Ar-sample cell. The slope of line 1 is caused by a change in the output power of the diode laser when its frequency is tuned by ramping of injection current. Line 2 is the absorption signal recorded by PD2 at the output of the discharge cell with argon. Line 3 is the signal from PD3 and corresponds to argon absorption in the reference cell.
Finally, line 4 in figure 2(a) shows the transmission resonances of a Fabry-Perot interferometer with a free spectral range of 750 MHz between the peaks. The distance between the peaks of the resonances determine the frequency range, which was used for a linearization procedure of the scale along the horizontal axis. After elimination of the baseline slope by dividing signal 1 to signal 2, followed by taking the logarithm the normalized spectral profile was obtained, as shown by points in figure 2 (b). These experimental points were fitted to the Voigt profile using the Origin software. In the range of gas pressures from 20 to 100 Torr and moderate temperatures, the $WD(T)$ and $WL(P,T)$ widths are comparable. In this case, fitting allows to determine values of $WD(T)$ and $WL(P,T)$ from the Voigt profile accurately.

Gas temperature $T$ averaged along the laser beam path was determined using expression (2) and the corresponding widths of the Doppler contours $WD(T)$. As it was shown in Ref. [6] the left part of relation (4) exhibits weak dependence on temperature gradient along the beam. The graph in figure 3(a) shows the temperature $T$ as a function of the gas pressure $P$ in the sample cell. It can be seen that the temperature of the gas varies from point to point. This is because during the pressure...
change in the sample cell it was necessary to adjust the matching circuit between the RF generator and the discharge. The calculated values also confirm that the temperature of neutral gas in the RF discharge is larger than room temperature (300 K). This is a convenient criterion for data verification.

The $WL(P,T)$ and $WD(T)$ widths make it possible, in accordance with formulas (2) and (4), to estimate the gas pressure $P_{\text{exp}}$ in the sample cell. If the values of $P_{\text{exp}}$ are put on a graph against the measured values of $P$ obtained from the pressure sensor Metran, then in the ideal case, the points must lie on the line with the unit slope coefficient and passing through the origin point of coordinate system (dotted line in figure 3(b)). This condition makes it possible to determine the value of the pressure broadening coefficient of the spectral line $\zeta(T_0)$. Since, if an incorrect $\zeta(T_0)$ value is used, then the points of the graph will not be approximated by a straight line that is in good agreement with the dotted line with a unit slope coefficient. In this work, the experimental points in figure 3(b) were fitted straight line with a slope coefficient equal to $1.00 \pm 0.06$, while the value of $\zeta(T_0)$ was chosen equal to $(2.4 \pm 0.5) \times 10^{-10}$ s\(^{-1}\)cm\(^3\). For comparison, the value of the same coefficient obtained by us in a somewhat different way [5, 6] was $\zeta(T_0)_{\text{Ar-Ar}} = (2.85 \pm 0.1) \times 10^{-10}$ s\(^{-1}\)cm\(^3\). Within the error limits, both values coincide.

Using the Doppler width $WD(T)$, it is possible to correct the temperature contribution to the shift $\Delta \nu$ of the spectral line. In accordance with (3), if the corrected shift $\Delta \nu$ is plotted along the vertical axis, and the gas pressure $P$ in the sample cell along the horizontal axis, a straight line should be obtained again. The experimental points are well approximated by a straight line as shown in figure 4. The error of the spectral line shift increases with pressure in the discharge. This is because the profile width of the investigated Ar line also increases with pressure in the discharge cell, which makes it difficult to determine the central frequency. However, the values of the slope coefficient defined either for the entire dataset or only for the points with small error are identical and equal to $-(6.7 \pm 0.2) \times 10^{-3}$ GHz/Torr. The value of the coefficient of pressure shift determined by the slope of the graph in figure 4 was $\gamma(T_0)_{\Delta \nu} = (2.1 \pm 0.1) \times 10^{-10}$ s\(^{-1}\)cm\(^3\). It is well known that the ratio $\gamma(T_0)/(2\zeta(T_0))$ must be equal to 0.368 [15]. The ratio of these coefficients defined in the work, gives a value of $0.44 \pm 0.09$, which is in good agreement with the theoretical estimate.

5. Conclusion
In this paper the method is proposed for simultaneous measurement of the pressure broadening and shift coefficients of the spectral lines, which makes it easy to take into account the contribution from the gas temperature. The broadening and shift coefficients for the 4s[3/2]\(_{2}\) \(\rightarrow\) 5p[3/2]\(_{3}\) Ar transition at 811.5 nm, which are important for the development of promising OPRGL lasers, were determined. Additionally it was demonstrated that easily accessible starters of fluorescent lamps can be used as sealed discharge cells for reference points of optical frequency.

Acknowledgments
This work was supported by the Ministry of Education and Science of the Russian Federation within the framework of the State Order under Project 3.1715.2017/4.6.

References
[1] Krupke W F 2012 Progress in Quantum Electronics 36 4
[2] Han J, Heaven M C 2012 Opt. Lett. 37 2157
[3] Han J, Glebov L, Venus G and Heaven M C 2013 Opt. Lett. 38 5458
[4] Demyanov A V, Kochetov I V and Mikheyev P A 2013 J. Phys. D: Appl. Phys. 46 375202
[5] Mikheyev P A, Chernyshov A K, Ufimtsev N I and Vorontsova E A 2015 Proc. SPIE 9255 92552W
[6] Mikheyev P A, Chernyshov A K, Ufimtsev N I, Vorontsova E A and Azyazov V N 2015 J. Quant. Spectrosc. Radiat. Transt. 146 1
[7] Gildina A R, Mikheyev P A, Chernyshov A K, Ufimtsev N I and Azyazov V N 2017
   *Photonics* 65 (5) 44
[8] Belostotskiy S G, Donnelly V M, Economou D J and Sadeghi N 2009
   *IEEE Trans. Plasma Sci.* 37 852
[9] Demtroder W 2003 *Laser Spectroscopy: basic concepts and instrumentation* (Berlin,
   Heidelberg, New York: Springer) chapter 3 pp 68–72
[10] Chernyshov A K, Chernyshova E A 2011 *Physics of Wave Phenomena* 19 89
[11] Chernyshov A K, Mikheyev P A 2016 *Proc. SPIE* 10176 101760T
[12] Chernyshov A K, Kotova S P 2006 *Instrum. Exp. Tech.* 49 92
[13] Sukenik C I, Busch H C 2002 *Rev. Sci. Instrum.* 73 493
[14] Chernyshov A K 2018 *Instrum. Exp. Tech.* 61 141
[15] Moussounda P S, Ranson P 1987 *J. Phys. B: At. Mol. Phys.* 20 949