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INVESTIGATION OF SPECTRAL LINE SHAPES BY INTRACAVITY LASER SPECTROSCOPY METHODS

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Abstract. Intracavity laser spectroscopy investigations of absorption line profiles in gas and plasma media are presented for study of some physical processes, that cause the profile transformation.

High-sensitive intracavity laser spectroscopy (ICLS) methods are widely applied for solving varied spectroscopic tasks. The main attention was given to determination of optical density and wavelengths of absorption. This allowed to find particle concentration and to identify a medium. Lesser attention was paid to spectral line contour measurements. However, such measurements can give additional important spectroscopic information which can be used for investigation of different physical processes in gaseous media.

Some aspects of application of ICLS methods for investigation of spectral line contours are developed in this paper.

Sufficiently exact measurement of optical density or amplification of atomic gaseous medium can be performed on varied frequencies by intracavity method, utilizing narrow-band laser. A magnetic field is convenient to scan the line contour about generation frequency. A measurement procedure is following. Initially, losses g_0 of laser cavity in the absence of the medium are determined at the frequency ν_0 by inserting additional losses upsetting the generation. Then, such additional losses g are measured in the presence of a medium. A difference $g_0 - g$ of losses gives a value of optical density (amplification) on the frequency ν_0 . Supplement of longitudinal magnetic field to medium in the case of simple Zeeman effect gives a possibility of measuring optical density (amplification) on the frequency ν_0 deturned from central frequency ν_0' of the contour by value $\Delta\nu = \nu_0' - \nu_0 + \Delta_H$, where Δ_H is magnetic splitting of line. So one can investigate spectral line contour by changing magnetic field intensity H.

The method described gives the most exact results when frequencies ν_0 and ν_0' coincide with each other and magnetic field does not influence the generated frequency and population of transition levels. As estimates show, systematic errors connected with above factors may be neglected, for example, when contours of 0.63 and 1.15 μm neon lines are investigating using helium-neon laser at $H < 400$ Oe.

Contours of both absorption of pure neon and amplification of helium-neon mixture at 1.15 μm wavelength are measured at different gas pressures, discharge currents and diameters of discharge tube. The contour of pure neon has Doppler form. In the case of helium-neon mixture a line broadening is determined mainly by Doppler effect, too. However, the contour form slightly differs from Doppler one. Moreover, a width of a line is increased comparetively to those in the case of pure neon under the same pressure and power, supplied to discharge. As investigations performed show, such effects are caused by transforming a part of potential energy into kinetic energy in result of second kind impacts.

The advantages of the ICLS for spectral lines profiles study are most completely realized when broadband tunable lasers are used. Plasma formations are highly perspective for such investigations. The spectrum recording is possible for a single microsecond or nanosecond laser pulse, that excludes distortions, caused by self-radiation of an object to be studied and its parameters temporal variation. The following such objects have been chosen :

1. The ablation plume, formed under laser radiation with flux power of $10^6 - 10^{10}$ W/cm^2 on a surface of aluminium target (Burakov *et al.*, 1995).

2. High current pulsed electric discharge in noble gases, nitrogen and carbon dioxide with strong erosion of transparent quartz wall (Burakov *et al.*, 1992).

3. The gas phase of a thermal graphite atomizer with barium, cesium and thallium vapours (Burakov *et al.*, 1994).

The instantaneous laser intensity in the modes with selective absorption is expressed by the following relationship similar to the Bouger-Lambert law :

$$i(\nu,t) = i'(t)exp[-k(\nu)ct\frac{L}{L_c}], \tag{1}$$

where $i'(t)$ is the instantaneous laser intensity beyond (near) the absorption line, $k(\nu)$ – the absorption coefficient at a frequency ν , c – the velocity of light, t – the current laser duration, L – the length of an absorbent in a cavity, L_c – the cavity base.

Experimentally, most often, measured are the integral values of the laser intensities $I(\nu)$ in the modes with absorption and I' near the line. As the absorption measure the value of relative intensity is used :

$$\frac{I(\nu)}{I'} = [1 + \frac{k(\nu)c\tau L}{nL_c(\frac{\tau}{t_m})}]^{-(n+1)}, \tag{2}$$

where τ is the total duration of laser pulse, t_m is the duration of laser pulse leading front, n is the parameter, characterizing the time shape of laser pulse ($n = 1$, 2 or 3). The relation (2) has been obtained for bell- shape time profile of laser pulse :

$$f(t) \sim (t/t_m)^n exp[-n(t/t_m - 1)]. \tag{3}$$

The spectral distribution $k(\nu-\nu_0)$ of an absorption coefficient (where ν_0 is the frequency of spectral line profile centre) can be found experimentally from the intracavity absorption spectrum with known time profile of a laser pulse. For the real in our experiments time profile with parameter n=2 :

$$k(\nu-\nu_0)=\frac{(I'/I)^{1/3}-1}{(I'/I_0)^{1/3}-1}, \tag{4}$$

where I_0 - integral laser intensitiy in the centre of absorption line at the frequency ν_0.

In ablation plasmas the Al I (λ = 394,4 nm) and Ca II (λ = 393,4 nm) spectral lines widths as well as their profiles have been measured for different moments of plume evolution (t) and various distances from the target surface (z). The maximal values of a width and aluminium line centre shift have been equal accordingly $\Delta\lambda$ = 0.22 nm and $\delta\lambda$ = 0.17 nm at the initial moments of a plume expansion (t = 1.2 μs and z = 1 mm). Under the same conditions the calcium line has $\Delta\lambda$ = 0.18 nm and $\delta\lambda$ = 0. These lines parameters provide determination of electrons density ($N = 0.9 \cdot 10^{18}$ cm^{-3}) and together with the species densities, measured from the lines intensities, a calculation of plasmas temperature (T = 2.3 eV) as well as plume pressure (P = 6 atm). At the late stages of plasmas expansion, including the afterglow, the mentioned lines parameters have been observed up to t = 80 μs and z = 5 mm.

In the plasmas of electric discharge the ICLS is an efficient method for component composition study, including the precise measurements of electrons density by using the width and profile of H_α line (hydrogen is a probe impurity in a gas).

The spectral lines profiles are possible to calculate and compare with experimental ones in a gas phase of thermal atomizer, taking into account Van-der-Vaals and Doppler broadening. The experimental (and calculated) widths of barium, cesium and thallium absorption lines are the following :

Ba I, 553.55 nm – 0.065 (0.041) nm,
Cs I, 455.54 nm – 0.090 (0.057) nm,
Tl I, 535.05 nm – 0.050 (0.041) nm.

The most satisfactory conformity of calculated and experimental data has been reached for thallium with resolved superfine structure , where the ratio of intensities of separate components has been measured - 1 : 0.36 and the distance between their centres - $1.08 \cdot 10^{-2}$ nm, while the calculated ones - 1 : 0.33 and $1.12 \cdot 10^{-2}$ nm, respectively.

There are situations when it is necessary to use a high optical density medium in laser cavity when ICLS method is applied. Such situation takes place, for example, in ICLS of two-photon transitions. Wide dips in intracavity absorption spectra appear in this case. Supplying of longitudinal magnetic field to medium in presence of polarizer in the cavity leads to sufficient transforming of dips. They are broadening and display one or more pairs of narrow resonances inside themselves. Components of each pair are situated symmetrically about central frequency ν_0 of transition. Investigations carried out allow to interpret these resonances as caused by rotation of polarization plane of light, because of Faraday effect. The condition of appearance of resonances is $\phi_\nu = n\pi/2$, where ϕ_ν is Faraday rotation angle at some frequency ν of absorbing contour, n is integer. This condition gives a possibility of measuring high optical densities by ICLS method, because of relation between volume ϕ_ν and optical density $k_0 l$ at the center of nonsplitted absorption contour. Such relation is expressed by proportional dependence $\phi_\nu = k_0 l F$, where F is a function of frequency deturning $\nu - \nu_0$, intensity H of magnetic field and line shape parameter $\Gamma/\Delta\nu_D$ (Γ is homogeneous width of line) (Voitovich, 1987). So, taking into account the condition of appearance of resonances, we have :

$$k_0 l = n\pi/2F. \tag{5}$$

The deturning $\nu - \nu_0$ can be determined as half difference of frequencies of resonances of the same pair with number n. One can simply calculate parameter $\Gamma/\Delta\nu_D$, if the temperature of gas is known. The value of H may be experimentally determined.

Optical densities of potassium vapour up to value $\sim 2 \cdot 10^5$ have been experimentally measured by described method. It is possible to find higher optical densities. Principally measurable value of $k_0 l$ is limited by the width of generated spectrum.

The results obtained indicate an efficiency of using of ICLS methods for investigation of spectral lines contours for purposes of plasma and gaseous media diagnostics.

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