

Dipole–dipole interactions between atoms in a partly excited resonance gas

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Abstract. In this paper, our experimental studies of dipole–dipole interactions in excited resonance gases are discussed. In weakly excited gas the dipole broadening of optical transitions is proportional to its density N . A gas medium can be identified by its density N as rarefied gas, where dipole broadening of atomic transitions is much less than Doppler width, or as dense gas, where dipole broadening is much more than Doppler width. According to our experiments the dipole broadening in rarefied gas does not depend on the optical excitation. This behavior is in a good agreement with predictions of standard theory for impact atomic collisions. In excited dense gas we observed the strong reduction of the dipole broadening. These experimental results are not described by standard theory for impact collisions. We suggested a simple description of the optical properties for excited dense gas.

Dipole–dipole interactions appear between identical quantum particles those are in the ground and the excited states. In a hot gas medium, the dipole–dipole interactions can be considered as particle’s collisions. These interactions can remarkably change the optical properties of resonance gases and plasmas.

One of the first theoretical analyses of collision processes was reported by Weisskopf 1933 [1]. The information about experimental and theoretical studies of dipole–dipole interactions in hot resonance gases is available in review papers [2–4].

Interaction potential W between atoms with dipole moments \vec{d}_1 and \vec{d}_2 and interatomic distance \vec{r} is described by the next expression [2]:

$$W = r^{-3}(-\vec{d}_1\vec{d}_2 + 3(\vec{n}\vec{d}_1)(\vec{n}\vec{d}_2)), \quad (1)$$

where \vec{n} is unit vector along vector \vec{r} . Spectral broadening of atomic transitions due to dipole–dipole interactions is defined by an average interaction potential: $\langle W \rangle = d^2N$, where N is the atomic density or number of atoms in cubic centimeter, d is an average dipole moment. The dipole–dipole potentials do not induce frequency shifts of atomic transitions [2]. The spectral dipole width Γ (FWHM) in weakly excited gas can be estimated by using the relation [2]:

$$\Gamma = KN = 2\alpha d^2 N/\hbar. \quad (2)$$

Here α is dimensionless parameter order of 1.



The properties of resonance gases depend on atomic density. We identify resonance gas by its density N as rarefied gas, where dipole broadening of atomic transitions Γ is much less than Doppler width $\Delta\omega_D$, or as dense gas, where dipole broadening Γ is much more than Doppler width $\Delta\omega_D$. It is convenient to use dimensionless Foigt parameter:

$$h = \Gamma/\Delta\omega_D. \quad (3)$$

When $h \ll 1$ the absorption profile of optically thin gas layer is inhomogenously broadened and described by the Gaussian function with Doppler width. When $h \gg 1$ the absorption profile of thin gas layer is homogenously broadened and described by Lorentz function with the dipole width Γ . In the intermediate case $h \sim 1$ the absorption profile of thin gas layer can be described by Foigt function. In the weakly excited gases (linear regime) the dipole–dipole interactions are studied quite well. In the most theoretical papers the dipole width is established as independent on excitation. Only in the theoretical work [5] the excitation dependence of the dipole broadening was analyzed in the frame of impact collisions. It was shown that in this model the dipole width in two-level atoms should be independent on the excitation. In literature there are not available information about experimental investigations of the dipole–dipole interactions in partly excited gases. Some our experimental studies of the dipole–dipole interactions at arbitrary optical excitation of rarefied gases [6–9] and dense gases [4, 10–13] were reported.

In our experiments we used the selective reflections from interface glass–gas as an effective spectroscopic technique which allowed us to study opaque gas media. Atomic metal vapors in the vacuum cells were served as resonance gas media. Spectral dependences of selective reflection are different for the rarefied and dense gases. Due to a spatial dispersion in the rarefied gas near a cell window a sub-Doppler structure appears in the selective reflection [14, 15]. In papers [16–18] it was suggested to apply selective reflection for Doppler-free spectroscopy of rarefied resonance gases. The derivative of sub-Doppler resonance, obtained by FM spectroscopic technique, is expressed as a dispersive Lorentzian function

$$dR/d\omega = A\Delta\omega(\Delta\omega^2 + \Delta\omega_{\text{SR}}^2)^{-1}. \quad (4)$$

Here A is a normalized factor, $\Delta\omega = (\omega - \omega_0)$ is the laser frequency detuning, $\Delta\omega_{\text{SR}}$ is sum of the natural width γ_{nat} and the dipole width Γ . By using the Doppler-free reflection spectroscopy the dipole widths of resonance transitions in cesium and rubidium were measured [7, 17, 19, 20]. This spectroscopic technique was applied to study an influence of optical saturation on dipole broadening [6–8]. The intensity dependence of the spectral width of atomic transitions in rarefied rubidium [7] and cesium [8] vapors can be extrapolated by expression

$$\Delta\omega_{\text{SR}}^2 = (\Gamma + \gamma_{\text{nat}})^2(1 + J/J_{\text{sat}}), \quad (5)$$

where J is intensity of the incident optical beam, J_{sat} is saturation intensity. Such behavior of the spectral width demonstrates excitation independence of the dipole broadening Γ in the rarefied gases. These observations support theoretical results of the work [5].

The optical properties of dense gases differ from the optical properties of rarefied gas. The optical susceptibility $\chi(\omega)$ of dense gas near resonance transition is expressed by Lorentz function [4]. The spectral profile of selective reflection coefficient can be calculated by using the dielectric function $\varepsilon(\omega) = 1 + 4\pi\chi(\omega)$ and Fresnel equation [4]:

$$R = |n - \sqrt{\varepsilon(\omega)}|^2 |n + \sqrt{\varepsilon(\omega)}|^{-2}, \quad (6)$$

where n is the refraction index of the cell window. From comparison of experimental and calculated (fitted) spectral profiles it is possible to determine spectral width and frequency shift

of resonance transitions. We are focused on study of partly excited dense gases. In paper [9] the first observation of reduction of dipole broadening and Lorentz shift in excited gas was reported. More accurate measurements were performed in the next works [4,10,13] by using the fit procedure with modified dielectric function which described below. The fit can be performed with a free parameter η which characterized the excitation level

$$\eta = (N_g - N_e)/N, \quad (7)$$

where N_g and N_e are the number densities of atoms in the ground and excited state. In the weakly excited gas η is practically equal to unit.

Our modified expression for the dielectric function $\varepsilon(\omega)$ is

$$\varepsilon(\omega) = 1 + KN\eta(\Delta\omega + \delta\omega_{\text{col}} + \eta\delta\omega_L + i(a + \eta b)\Gamma/2)^{-1}. \quad (8)$$

Here $\delta\omega_{\text{col}}$ is excitation independent frequency shift, $\eta\delta\omega_L = \eta KN/6$ is Lorentz local field correction, $\Gamma(\eta) = (a + \eta b)\Gamma = (a + \eta b)KN$ is excitation dependent dipole broadening. By taking into account the redistribution atoms between excited states $P_{1/2}$ and $P_{3/2}$ in the potassium and rubidium metal vapors we obtain the next relation $\Gamma(\eta) = (a + \eta b)KN = KN_g$ [4, 9, 10]. The dipole broadening in the excited atomic vapor is proportional to the ground state population. This experimental result can be explained by very weak interactions between excited atoms in dense gases to (like van der Waals interactions) to compare with strong dipole interactions between excited and ground state atoms. The detailed theoretical analysis of excited resonance gas media will be appreciated in order to clarify the situation.

In conclusion, we would like to say that our results can be used to describe the optical properties of excited resonance gases for densities where Voigt parameter $h \ll 1$ and $h \gg 1$.

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