Nonlinear reflection of laser beam from high density atomic vapor

V A Sautenkov\textsuperscript{1,2}, S A Saakyan\textsuperscript{1}, E V Vilshanskaya\textsuperscript{1,3}, M A Gubin\textsuperscript{2}, B B Zelener\textsuperscript{1,3,4} and B V Zelener\textsuperscript{1}

\textsuperscript{1} Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
\textsuperscript{2} Lebedev Physical Institute of the Russian Academy of Sciences, Leninsky Avenue 53, Moscow 119991, Russia
\textsuperscript{3} National Research University Moscow Power Engineering Institute, Krasnokazarmennaya 14, Moscow 111250, Russia
\textsuperscript{4} National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe Shosse 31, Moscow 115409, Russia

E-mail: vsautenkov@gmail.com

Abstract. We have investigated the nonlinear selective reflection of a laser beam (\(\lambda = 780\) nm) from a high density rubidium vapor. The laser frequency was tuned over atomic transitions and coherent optical excitation of the atomic vapor was realized. The observed narrowing of reflection spectral profile is associated with a competition of two nonlinear processes: optical field induced coherent broadening (Rabi broadening) and optical saturation induced decreasing of dipole–dipole broadening of atomic transitions. For more complete understanding the spectral narrowing we suggest to perform a set of new experiments.

1. Introduction
Our goal is to study nonlinear optical processes in atomic vapors in presence of dipole–dipole interactions. The dipole–dipole broadening of resonance transitions in a weakly excited atomic vapor can be expressed as [1]:

\[
\Gamma = K 2\pi \left( \frac{\lambda}{2\pi} \right)^3 N \left( \frac{2J_e + 1}{2J_g + 1} \right)^{1/2} \gamma_n,
\]  

(1)

where symbol \(\lambda\) is presented a resonance wavelength, \(N\) is an atomic number density, \(\gamma_n\) is the natural linewidth of the resonance transition, and factor \(K \approx 1\). The atomic vapors can be considered as dense resonance gases when the atomic number density \(N\) is more than \((2\pi/\lambda)^3\). Under these conditions the collective atom–light interactions can be observed [2].

Taking into account a value of broadening \(\Gamma_{\text{trans}}\) for transitional atomic number density \(N_{\text{trans}} = (2\pi/\lambda)^3\) and then comparing dipole–dipole broadening \(\Gamma\) with Doppler broadening \(\Delta \omega_D\) it is possible to introduce the next useful definitions:

- low density atomic vapor (dilute vapor), where \(\Gamma < \Gamma_{\text{trans}}\) and \(\Gamma \ll \Delta \omega_D\);
- dense atomic vapor (intermediate density), where \(\Gamma_{\text{trans}} \leq \Gamma \leq \Delta \omega_D\);
- high density atomic vapor, where \(\Gamma > \Delta \omega_D\).
In dilute atomic vapors under conditions of a strong optical saturation a variation of the dipole–dipole broadening was not observed \cite{3,4}. Obtained experimental data are in agreement with results of theoretical work \cite{5} where the excitation independence for the dipole–dipole broadening was predicted for dilute gases in the model of the impact collisions. In experiments with dense atomic vapor (intermediate density) \cite{6} and high density atomic vapors \cite{7–9} a linear relations between dipole–dipole broadening and excitation level of atomic transitions were observed and analyzed. The change of the spectral width is associated with quasistatic behavior of dipole–dipole interactions at the high densities \cite{7}. We shall note that in the described experiments \cite{6–9} the pump-probe scheme was used and non-coherent optical excitation of atoms was realized. The pump laser frequency was detuned far away from the atomic resonance and the coherent optical field broadening could be neglected. In present paper the preliminary observations of selective reflection profiles with coherent optical excitation of atomic transitions in high density vapor is discussed.

2. Experimental results and discussion

In our experimental research of the natural abundance of isotopes $^{85}\text{Rb}$ and $^{87}\text{Rb}$ was used. The rubidium atomic vapor was confined in optical cell ($L = 10$ cm) which described in \cite{10}. This cell has garnet windows and can be heated up to 400 $^\circ$C. The atomic number density of saturated rubidium vapor was defined by temperature of coldest sport of the cell. The vapor cell at different temperatures was used for observation of reflection and absorption spectra at hyperfine components of $D_2$-line of $^{85}\text{Rb}$ and $^{87}\text{Rb}$ atoms ($\lambda = 780$ nm).

As source of tunable monochromatic light we applied the commercial external cavity laser with amplifier (Toptica Photonics). The output laser power can increase up to 2 W and laser linewidth was less than 0.5 MHz. The laser frequency was tuned over hyperfine components of $D_2$-line of rubidium. In order to get effective optical saturation of the selective reflection the laser beam was focused on internal surface of the front window of the cell. The area of the light beam sport on interface window/vapor was order $2 \times 10^{-4}$ cm$^2$. Linear and nonlinear reflection spectra are presented in figure 1(a, b). At the working atomic number density $N = 3.3 \times 10^{16}$ cm$^{-3}$ the dipole–dipole broadening of hyperfine components of $D_2$-line $\Gamma/2\pi$ is equal to 3.3 GHz (the broadening factor $\Gamma/N$ is equal to $2\pi \times 10^{16}$ cm$^3$/s \cite{1,2}).

It is clear that the spectral profile of the nonlinear reflection in figure 1(a) is narrower than the spectral profile of the linear reflection in figure 1(b). Also in the nonlinear spectrum new features related with hyperfine splitting of the ground states of $^{85}\text{Rb}$ and $^{87}\text{Rb}$ atoms. The spectral narrowing under conditions of the coherent optical excitation is observed. We would like to remind that optical excitation can be called coherent excitation when the value of Raby frequency is comparable or more than relaxation rate of the optical coherence.

It is interesting to compare presented observations with results obtained in \cite{11} with high density rubidium vapor under experimental condition for non-coherent optical excitation in pump-probe scheme with large detuning of pump laser frequency. Some the main experimental parameters in work \cite{11} are quite close to the similar experimental parameters in the presented research. For example, the atomic number density of rubidium vapor $N$ was of $3.5 \times 10^{16}$ cm$^{-3}$, the area of the pump beam spot on interface window/vapor was order of $10^{-4}$ cm$^2$, power of pump beam was 0.12 W. Anyway the narrowing of the spectral profile of the nonlinear reflection in the work \cite{11} is clearly more pronounced than in figure 1 above. It can be explained by the published statement \cite{7–9} that in non-coherent excitation process the reduction of the dipole–dipole broadening depends only on the difference between the ground state and excited state populations. In this case the coherent optical field broadening can be neglected. Under condition of coherent optical excitation there is a competition between reduction dipole–dipole broadening related with vapor excitation and the optical field broadening proportional to the Rabi frequency (Rabi broadening). In our case the Rabi frequency $\Omega/2\pi$ is order of 2.5 GHz. With such large
Figure 1. (a) Linear selective reflection of laser beam with low power $P = 5 \text{ mW}$. (b) Nonlinear selective reflection of laser beam with high power $P = 0.2 \text{ W}$. (c) Linear absorption spectra for the rubidium vapor in the cell at room temperature. The atomic number density of rubidium vapor $N$ was of $3.3 \times 10^{16}$ (a, b) and about $10^{11} \text{ cm}^{-3}$ (c).

value of Raby frequency it was un-expectable to observe the spectral narrowing in the real experiments.

In order to get detailed description of the spectral narrowing in coherently excited resonance media it is necessary to perform additional experiments with nonlinear selective reflection at different atomic densities and make careful theoretical analysis. Supplementary important information can be obtained by using the nanocells where dipole broadened absorption spectra for high density vapors can be recorded directly [12].

3. Conclusions
The nonlinear selective reflection of a laser beam from dense rubidium vapor was studied under condition of a coherent excitation. The pronounced narrowing of the reflection profile due to optical saturation was observed. The effect was explained by optical saturation induced reduction of dipole–dipole broadening of atomic transitions. Contribution of coherent field broadening was relatively small to compare with the optically induced decreasing of dipole–dipole broadening. The obtained results can help to understand of new nonlinear effects in dense resonance gases.

Acknowledgments
We thank M A Kazaryan (Lebedev Physical Institute RAS, Moscow, Russia) and M N Shneider (Princeton University, Princeton, USA) for useful discussions. The work was supported by the Presidium RAS (basic research program “Investigation of Matter in Extreme States” headed by V E Fortov).
References

[1] Lewis E L 1980 Phys. Rep. 58 1–71
[2] Keaveney J 2014 Collective Atom-Light Interactions in Dense Atomic Vapours Springer Theses (Durham: Springer International Publishing)

[3] Vuletic V, Sautenkov V A, Zimmermann C and Hänsch T W 1994 Opt. Commun. 108 77–83
[4] Sautenkov V A, Saakyan S A, Bronin S Ya, Klyarfeld A B, Zelener B B and Zelener B V 2018 J. Phys.: Conf. Ser. 946 012127

[5] Vdovin Y A and Dobrodeev N A 1969 Sov. Phys. JETP 28 544–8
[6] Meng T F, Ji Z H, Zhao Y T, Xiao L T and Jia S T 2016 Chin. Phys. Lett. 33 113202
[7] Sautenkov V A, Van Kampen H, Eliei E R and Woerdman J P 1996 Phys. Rev. Lett. 77 3327–30
[8] Li H, Sautenkov V A, Rostovtsev Y V and Scully M O 2009 J. Phys. B: At. Mol. Opt. Phys. 42 065203

[9] Sautenkov V A 2011 Laser Phys. Lett. 8 771–81

[10] Sautenkov V A, Varzhapetyan T S, Li H, Sarkisyan D and Scully M O 2010 J. Russ. Laser Res. 31 270–5
[11] Li H, Varzhapetyan T S, Sautenkov V A, Rostovtsev Y V, Chen H, Sarkisyan D and Scully M O 2008 Appl. Phys. B: Lasers Opt. 91 229–31

[12] Varzhapetyan T, Nersisyan A, Babushkin V, Sarkisyan D, Vdovic S and Pichler G 2008 J. Phys B: At. Mol. Opt. Phys. 41 185004