Electro-optical effects in dense and cold atomic gases

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On the basis of the general theoretical approach developed previously by Fofanov *et al.* [Ya. A. Fofanov *et al.*, Phys. Rev. A **84**, 053811 (2011)], we analyze the atomic polarization created by weak monochromatic light in an optically thick, dense, and cold atomic ensemble placed in an electrostatic field. On this basis, we determine complex refractive indexes and the permittivity tensor of the medium. We analyze the dispersion of the permittivity and its dependence on the strength of the field and on the density of the medium. We show that electro-optic effects in dilute and cold gases differ essentially from those in dense ones. In the latter case, electric field modifies the shape, amplitude, and typical linewidth of the atomic resonance. The electric field also modifies the collective Lamb shift. Observed peculiarities are explained as a result of the indirect influence of the field on optical properties of dense gases through modification of polyatomic collective effects caused by interatomic dipole-dipole interactions.

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I. INTRODUCTION

Improvements in techniques for cooling atomic gases in atomic traps make their utilization very promising for applications in various areas of fundamental science and technology such as metrology, the development of frequency standards, and quantum information problems [1–7].

Very recently, special attention has been focused on dense ensembles, in which the mean free path of resonant photons is comparable with their wavelength. This attention is partially due to the possibility to create atomic clouds with a very large optical thickness, which is important for almost all of the above-mentioned applications. Besides that, such ensembles exhibit a number of unique physical properties, such as subradiance, a collective Lamb shift, cold-atom-based random lasing, and possible strong localization of light, which are either fundamentally impossible or suppressed in low-density media (see [8–18] and references therein).

The efficiency of cold atom applications and possibilities to observe one or another physical phenomenon are determined to a great extent by the possibilities to prepare an exploitable ensemble with appropriate properties. Often the control over these properties in real time is required. One of the most simple and effective methods of modification of physical, particularly optical, properties of cold atomic samples is based on the application of an external magnetostatic or electrostatic field.

Magneto-optical and electro-optical effects have been known for a long time and, by now, have been studied in detail for various materials. In particular, some aspects of the influence of a magnetic field on optical properties of cold atomic ensembles have been analyzed both experimentally and theoretically. Experiment [19] revealed a very large Verdet constant for dilute atomic clouds. In these works [20–22], an unusual influence of a magnetic field on coherent backscattering was discovered. It was shown that contrary to expectation, a magnetic field can increase the enhancement of backscattering in some cases. The effects of this field on the resonant dipole-dipole interatomic interaction and multiple recurrent scattering in dense and cold atomic ensembles were investigated in detail in [14,23,24]. The role of the electrostatic field in the case of cold atomic clouds has been studied in much less detail.

The principal goal of the present paper is to analyze theoretically the influence of electric fields on optical properties of nondegenerate cold atomic ensembles. We will focus our attention on the case of dense gases. There is a good reason to believe (see [25]) that electro-optical effects in such gases differ essentially from those in dilute ones. We will study how the dielectric susceptibility tensor of the system changes with the density of atoms and with the strength of electric field.

Our analysis is based on the quantum microscopic approach suggested in [26,27] for examination of coherent and incoherent light transport in the case of dense gases when the Lorentz-Lorenz local field theory [28] cannot be applied. This approach assumes the study of the spatial distribution of atomic excitation and atomic polarization created in the gases by weak monochromatic light. On this ground, we calculate a numerically complex index of refraction and dielectric constant of the medium for different frequencies of the incident light.

We will show that the electrostatic field strongly influences the nature and manifestation of electro-optical effects in dense gases. Particularly in contrast with the case of dilute gases, for dense ones the electric field modifies all characteristics of the atomic susceptibility. It changes the shape, amplitude, and typical linewidth of the atomic resonance. The electric field also modifies the collective Lamb shift observed in dense atomic ensembles.

II. BASIC ASSUMPTIONS AND APPROACH

In the present work, we analyze the steady state of an atomic ensemble consisting of N atoms interacting with a quasiresonant monochromatic electromagnetic field and

vacuum modes. These atoms are identical and have ground state $J = 0$ separated by the frequency ω_a from the excited $J = 1$ state. The natural linewidth of the three Zeeman sublevels of this state ($m = -1,0,1$) are γ. Atoms are assumed to be motionless. We consider a statistical ensemble of clouds with the random distribution of atoms. The main results presented below are obtained by averaging over this ensemble by the Monte Carlo method. The external quasiresonant light is a plane monochromatic wave with frequency ω_s,

$$\mathbf{E}_l = \mathbf{e}E_0 \exp(-i\omega_s t + i\mathbf{kr}). \tag{1}$$

Here, $\mathbf{e}$ and E_0 are the polarization vector and amplitude of the field. The intensity of the radiation is assumed to be sufficiently small that all nonlinear effects are considered negligible.

At present, there are several similar approaches to the description of weak light interaction with dense and cold atomic clouds [29–39]. Our theoretical analysis in this work is based on the consistent quantum-posed theoretical approach developed previously in [26,36]. According to [26,36], under the considered conditions, the nature of atomic excitation is determined by the amplitudes of collective atomic states with one excited atom b_e while the other atoms are in the ground state. Here the index e contains information about both the number of the excited atom and specific Zeeman sublevel that is populated.

Considering weak coherent light as a superposition of a vacuum state and a small admixture of a one-photon Fock state, for the Fourier components $b_e(\omega)$ of these amplitudes we obtain the following set of coupled-dipole equations (for greater length, see [36]):

$$\sum_{e'}[(\omega - \omega_a)\delta_{ee'} - \Sigma_{ee'}(\omega)]b_{e'}(\omega) = V_e(\omega). \tag{2}$$

Here, matrix $\Sigma_{ee'}(\omega)$ describes the excitation exchange between different atoms and spontaneous decay of each atom. In the case when the atoms are placed in an external electric field, it also takes into account the Stark frequency shift caused by this field. In this case, in the reference frame with the quantization axis directed along the electric field, the matrix $\Sigma_{ee'}(\omega)$ has the form

$$\begin{aligned}\Sigma_{ee'}(\omega) = {} & -\gamma/2(i - \Delta)\delta_{ee'} + (1 - \delta_{ee'}) \\ & \times \sum_{\mu,\nu} \frac{\mathbf{d}^{\mu}_{eg}\mathbf{d}^{\nu}_{g'e'}}{\hbar r^3} \exp\left(i\frac{\omega_a r}{c}\right) \\ & \times \left\{\delta_{\mu\nu}\left[1 - i\frac{\omega_a r}{c} - \left(\frac{\omega_a r}{c}\right)^2\right]\right. \\ & \left. - \frac{\mathbf{r}_\mu \mathbf{r}_\nu}{r^2}\left[3 - 3i\frac{\omega_a r}{c} - \left(\frac{\omega_a r}{c}\right)^2\right]\right\}. \end{aligned} \tag{3}$$

Here, $\Delta = 2\Delta\omega^S_{eg}/\gamma$ is the Stark shift of the atomic transition $e \leftrightarrow g$ measured in units of the natural half width of the line (note that in the considered case of two-level atoms, the electric field shifts the level in such a way that the frequency of the transition $J = 0,m = 0 \leftrightarrow J = 1,m = 0$ is always more than for $J = 0,m = 0 \leftrightarrow J = 1,m = \pm 1$ and the corresponding normalized shift Δ is positive); $\mathbf{r}_\mu$ is the projection of the vector $\mathbf{r}$ on the axis of the chosen coordinate system and $r = |\mathbf{r}|$ is the separation between the atoms excited to the states e and e'.

Vector $V_e(\omega)$ on the right-hand side of Eq. (2) describes the excitation of cloud atoms by the external radiation. It is determined by the initial excitation through the nonmodified external field

$$V_e(\omega) = 2\pi\delta(\omega - \omega_s)V_{e0}; \tag{4}$$

$$V_{e0} = -\frac{\mathbf{d}_{eg}\mathbf{e}E_0 \exp(i\mathbf{kr}_e)}{\hbar}, \tag{5}$$

where $\mathbf{d}_{eg}$ is the dipole matrix element for transitions from the ground g to the excited e state of the atom and $\mathbf{r}_e$ is the radius vector of the atom excited in the state e.

Introducing an inverse matrix for the system (2),

$$R_{ee'}(\omega) = [(\omega - \omega_a)\delta_{ee'} - \Sigma_{ee'}(\omega)]^{-1}, \tag{6}$$

we can write its formal solution as follows:

$$b_e(\omega) = \sum_{e'} R_{ee'}(\omega)V_{e'}(\omega). \tag{7}$$

In the present work, we determine the matrix $R_{ee'}(\omega)$ and $b_e(\omega)$ numerically.

For time-dependent amplitude $b_e(t)$, we get

$$b_e(t) = \exp(-i\omega_s t)\sum_{e'} R_{ee'}(\omega_s)V_{e'0}. \tag{8}$$

Knowing the amplitudes $b_e(t)$ allows us to calculate the atomic polarization as the total dipole moment per unit volume of the cloud. In analyzing this polarization, it is convenient to select positive- and negative-frequency parts and use a basis of circular polarization ($\mu = 0, \pm 1$),

$$\mathcal{P}_\mu(\mathbf{r},t) = \mathcal{P}^{(-)}_\mu(\mathbf{r},t) + \mathcal{P}^{(+)}_\mu(\mathbf{r},t). \tag{9}$$

In the steady-state condition, the time dependence of the complex atomic polarization is given by the simple exponential function

$$\mathcal{P}^{(+)}_\mu(\mathbf{r},t) = \mathcal{P}^{(+)}_\mu(\mathbf{r})\exp(-i\omega_s t). \tag{10}$$

For the spatial distribution of the amplitude $\mathcal{P}^{(+)}_\mu(\mathbf{r})$ from (8), we find

$$\mathcal{P}^{(+)}_\mu(\mathbf{r}) = \frac{1}{\Delta V}\sum_{a\in\Delta V}\sum_{e'} R_{e^m_a e'}(\omega_s)V_{e'0}. \tag{11}$$

Here we added additional indexes m and a at e to explicitly show that under summation, we have to include only atoms a located in a mesoscopic volume ΔV and only those states e^m_a of these atoms which give a contribution to the corresponding projection of the polarization vector. In the basis of circular polarization, such a contribution comes only from one Zeeman sublevel with $m = \mu$. Note, however, that due to optical anisotropy of the atomic ensemble caused by the electrostatic field $\mathbf{E}$, orientation of the atomic polarization vector in the general case does not coincide with orientation of the polarization of light exciting the atoms.

In the next section, we will use relation (11) to calculate the spatial distribution of atomic polarization and to analyze on this background the influence of an external electrostatic field on coherent light transport in ensembles of different densities.

III. RESULTS AND DISCUSSION

For the purpose of this work, we consider cylinder-shaped atomic ensembles with a radius R and length L. The random distribution of atoms is assumed to be uniform, on average. We will also assume that this ensemble is irradiated from one end by a monochromatic plane wave with the wave vector directed along the axis of the cylinder.

To find two principal values of the permittivity tensor, we analyze two polarization schemes, in which the direction of the induced atomic polarization coincides with the polarization of the exciting radiation. In the first case, external quasiresonant light is circularly polarized and the electrostatic field is directed along the axis of the cylinder $\mathbf{E}\|\mathbf{k}$. The second case corresponds to radiation linearly polarized along the electrostatic field, which is perpendicular to the axis of the considered cylindrical volume $\mathbf{E}\perp\mathbf{k}$. In both cases, the polarization of the external radiation does not change under its propagation in the atomic ensemble.

In Fig. 1, we show the spatial dependence of the absolute value [Fig. 1(a)] and phase [Fig. 1(b)] of $\mathcal{P}^{(+)}(\mathbf{r})$ for different polarization schemes and different detuning δ of exciting radiation from the resonance frequency of the corresponding atomic transition. The calculations were made for a cloud with length $L = 10$ and radius $R = 20$ (hereafter, in this paper, we use the inverse wave number of the probe radiation in vacuum, $k_0^{-1} = \lambda/2\pi$, as the unit of length). The average density of atoms is $n = 0.15$.

To avoid the influence of boundary effects at the lateral surface of the cylinder, we calculate the atomic polarization for a zone near the axis of the cylinder where the dependence of the polarization on r is negligible. In this area, the polarization depends only on z. Our analysis shows that for the considered parameters, this takes place for the inner part of the cylinder with $r \leqslant 12$. The results shown in Fig. 1 are obtained by averaging of the atomic polarization over a region with radius $r = 8$.

Figure 1 shows that outside the boundary region near $z = 0$, the phase of the complex polarization begins to increase linearly and the amplitude of the polarization decreases exponentially with z. As the depth z increases, we see some deviations from such behavior. For detuning $\delta = 0$, these deviations take place practically only in the boundary area near the far end of the cylinder ($z = 10$). For $\delta = 0.3\gamma$, the nonlinear behavior of curves in Fig. 1 can already be seen at $z \geqslant 5$.

Our calculation shows that the size of the region where the curves in Fig. 1 are linear increases monotonically as we increase the number of statistical tests. The size also decreases as the frequency ω_s approaches the resonance frequency of the considered dense media. It gives us a good reason to believe that deviation from linearity is the result of statistical errors caused by the fact that far from $z = 0$, the averaged polarization is extremely small in comparison with the polarization corresponding to any random specific spatial configuration of the ensemble. So here we have to calculate the numerically small difference of big numbers. The curves in Fig. 1 were obtained by averaging over more than 5×10^5 different random spatial configurations of the atomic ensemble. They were not additionally smoothed. In spite of such a huge number of statistical tests, the curves contain the noticeable traces of fluctuations for big z.

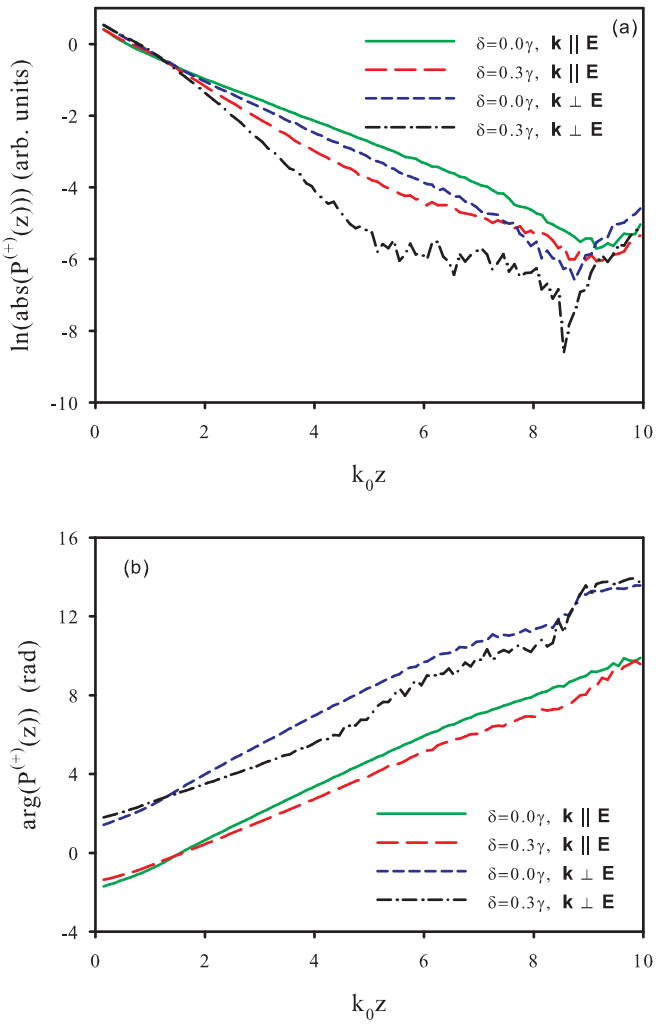

FIG. 1. Spatial distribution of atomic polarization for two principal polarization schemes. (a) Natural logarithm of amplitude of polarization, $\ln\{\mathrm{abs}[\mathcal{P}^{(+)}(z)]\}$; (b) phase of polarization, $\arg[\mathcal{P}^{(+)}(z)]$. Calculations were made for a cylindrical cloud with length $L = 10$ and radius $R = 20$; atomic density is $n = 0.15$; $k_0 = \omega_s/c$ is the wave number of the source radiation in vacuum. The detuning δ is calculated from the resonance frequency of the corresponding transition.

Further, all conclusions about atomic susceptibility will be made on the basis of analysis of linear sections of curves $\ln\{\mathrm{abs}[\mathcal{P}^{(+)}(z)]\}$ and $\arg[\mathcal{P}^{(+)}(z)]$, i.e., for those spatial intervals where atomic polarization can be presented as a simple exponential function, $\mathcal{P}^{(+)}(z) = \mathcal{P}_0 \exp[i(k' + ik'')z]$.

For regions away from the boundaries, we deal with a quasiuniform medium. In such a case, the coordinate dependence of the electric field inside the medium $\mathcal{E}^{(+)}(z)$ is the same as of $\mathcal{P}^{(+)}(z)$, with the same coefficients k' and k'', i.e., $\mathcal{E}^{(+)}(z) = \mathcal{E}_0 \exp[i(k' + ik'')z]$. So knowing the polarization of the atomic ensemble allows one to make some conclusions about light propagation in it, particularly about the imaginary k'' and real k' parts of the complex wave number of the plane electromagnetic wave in the atomic ensemble. The quantities

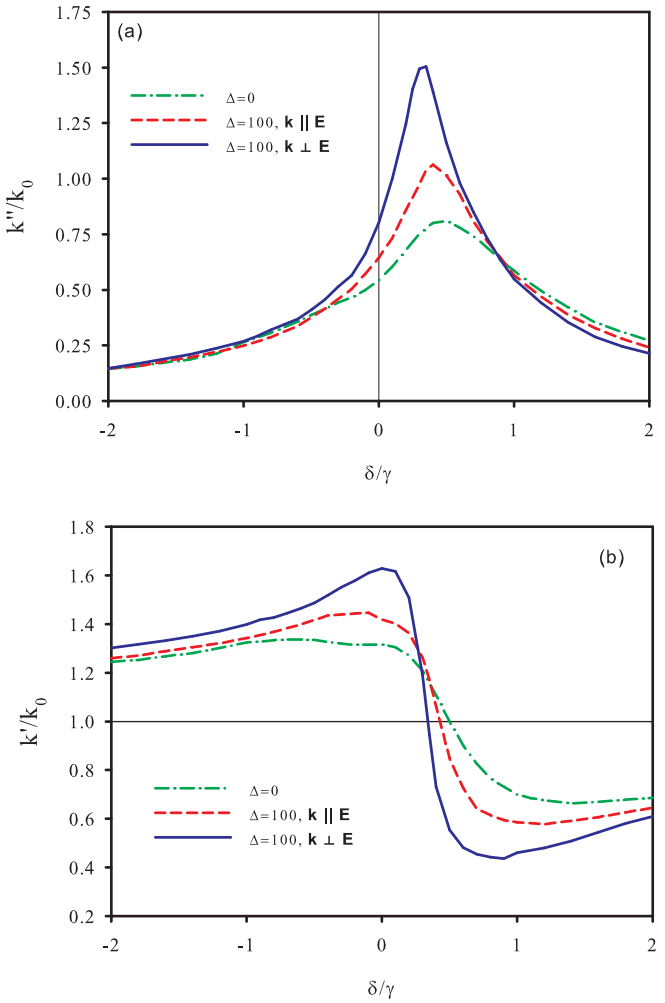

FIG. 2. Spectrum of (a) imaginary and (b) real parts of the complex wave number of the electromagnetic wave. Atomic density is $n = 0.2$. The detuning δ is calculated from the resonance frequency of the corresponding transition.

k'' and k' in turn give information about the refractive index and absorption coefficient of the media under consideration.

We performed the calculations of the spatial distribution of atomic polarization for a wide range of conditions, particularly for different frequencies of exciting radiation and for different densities of the atomic ensemble. It gives us an opportunity to analyze the coherent light propagation in various conditions.

Figure 2 illustrates the influence of a strong electrostatic field on the spectral dependence of refraction and absorption of probe radiation in dense and cold atomic gases. The results for the two main polarization schemes are compared here with the case when the electric field is absent ($\Delta = 0$). The calculation is made for $n = 0.2$. To have the possibility to compare the shape of the spectral dependencies, we measure detuning δ from the resonance frequency of the corresponding transition. For the case $\mathbf{E}\perp\mathbf{k}$, it is the $J = 0, m = 0 \leftrightarrow J = 1, m = 0$ transition. In the case of circularly polarized exciting radiation propagated along electrostatic field $\mathbf{E}\|\mathbf{k}$, the detuning is counted off $J = 0, m = 0 \leftrightarrow J = 1, m = \pm 1$ atomic frequency.

Figure 2 demonstrates the fundamental differences of electro-optical effects in dense gases as compared with dilute ones.

The properties of dilute atomic ensembles can be described on the basis of a one-body density matrix. The main components of the one-atom polarizability tensor are modified by an external electrostatic field, but its influence is restricted by the Stark shift of the resonant frequencies. The amplitudes, widths, and shape of the resonances themselves remain unchanged. As we can see from Fig. 2, for dense cold gases it is not the case. The electrostatic field changes all the characteristics of the atomic resonance. Comparison of the dash-dotted curve with the others shows that the shape, amplitude, and typical linewidth of the atomic resonance change in the electric field. Moreover, we see that the electric field modifies the collective Lamb shift. It becomes noticeably less.

The influence of the field manifests itself differently for the two main polarization channels. This influence is more pronounced at the transition $J = 0, m = 0 \leftrightarrow J = 1, m = 0$ (see solid line in Fig. 2). For the considered parameters, the absorbance at this transition increases by more than 85%. For the circularly polarized light propagating along the electrostatic field $\mathbf{E}\|\mathbf{k}$, the electro-optical effects are a bit less distinct (dashed lines in Fig. 2), but they are not small in this case either.

The discovered peculiarities of the electro-optical effects in dense gases can be accounted for by a specific mechanism of the influence of the electrostatic field. The Stark shift in dense ensembles affects not only the susceptibility of individual atoms but also additionally modifies the resonant dipole-dipole interatomic interaction. As is known, this interaction can be explained by the exchange of virtual photons between the atoms and the shifts of atomic levels caused by the electrostatic field modifying this exchange. Thus, this field affects the nature of the collective polyatomic effects. Such an indirect influence on optical properties through modification of near-field effects and interatomic correlations can be much more effective than the direct influence observable in single-atom effects.

Such character of electro-optical effects in a dense atomic ensemble means that the influence of an electrostatic field will change with the density. In Fig. 3, we illustrate how the optical properties of an atomic ensemble placed in an electric field depend on its density n. We consider this dependence by the example of one of the main components of the permittivity tensor. This main component corresponds to linearly polarized light propagating perpendicularly to a constant field $\mathbf{E}\perp\mathbf{k}$. The calculation is performed for strong electric field $\Delta = 100$.

Figure 3 demonstrates the expected qualitative behavior of the dielectric constant. The amplitude of the resonance [Fig. 3(b)] increases with density as well as its shift and width. In increasing of the amplitude, there is a tendency towards saturation. Similar density dependence is observed in the case of zero field (see [26,40]); however, there are essential quantitative differences. The electric field noticeably suppresses the saturation effect.

To keep the picture clear, we do not show curves for the zero electric field in Fig. 3. But we can illustrate this suppression by some numerical examples. Density increasing from $n = 0.1$ to $n = 0.4$ leads in the considered case to increasing of the imaginary part of ε by the factor close to 3, while in the case of zero field, the corresponding increase is only 75%.

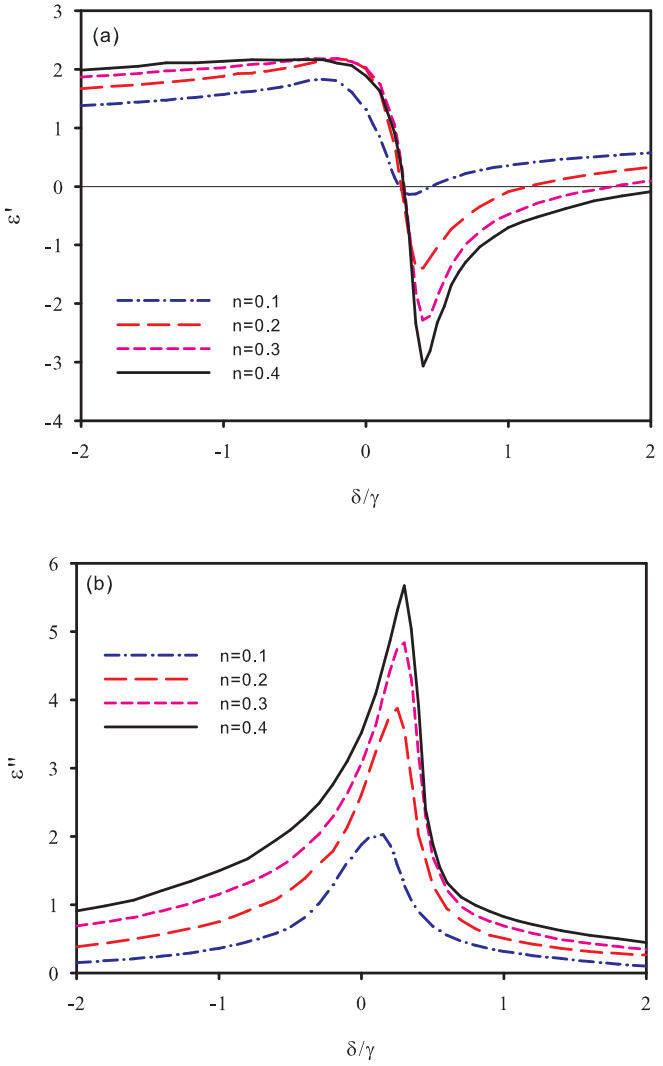

FIG. 3. Spectrum of (a) real and (b) imaginary parts of one of the main components of the permittivity tensor for different densities n. Polarization scheme is $\mathbf{E}\perp\mathbf{k}$, $\Delta = 100$.

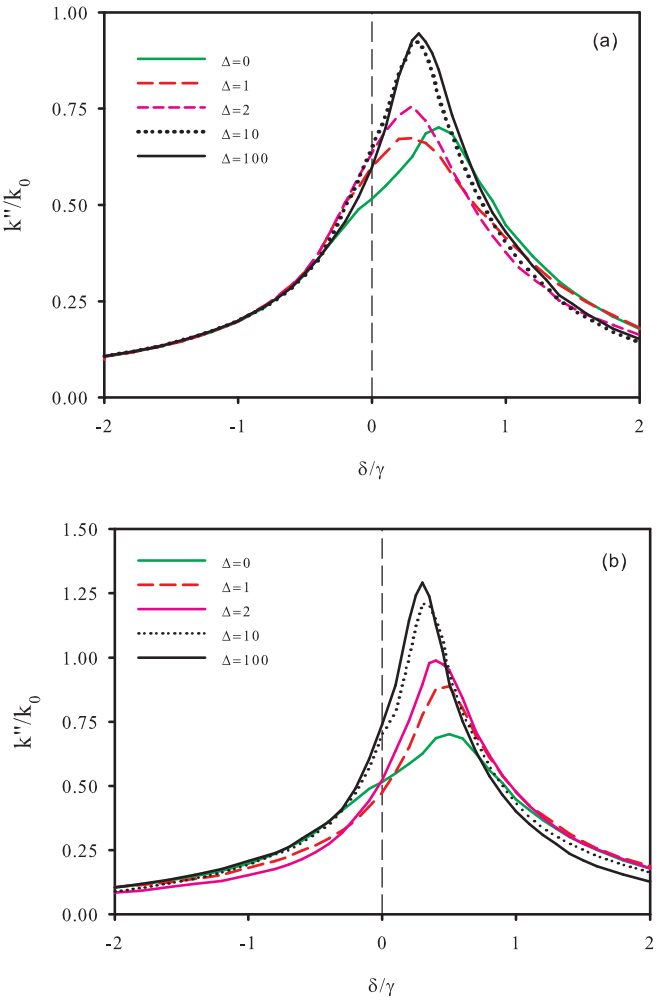

FIG. 4. Spectrum of the imaginary part of one of the main components of the wave number for different electrostatic field strength and for two main polarization schemes (a) $\mathbf{E}\|\mathbf{k}$ and (b) $\mathbf{E}\perp\mathbf{k}$. The detuning δ is calculated from the resonance frequency of the corresponding transition. Atomic density is $n = 0.15$.

The saturation plays a negative role because it does not allow one to obtain atomic ensembles with very big optical depth just by increasing the density. As we mentioned above, the possibilities to prepare atomic samples with big optical thickness are important for various practical applications of cold gases and an electric field can partially help to solve this problem.

The suppression of saturation effects in an electric field is accompanied by the narrowing of resonance curves compared with the case of zero field. It can be seen from Fig. 2, but for bigger densities narrowing becomes more pronounced. Thus our calculations show that for $n = 0.3$, a strong electric field decreases the full width at half maximum more than three times. It means that as the atomic density increases, the modification of the optical parameters of the atomic ensemble in the given electric field becomes more considerable. It can also be seen from Fig. 3(b) that the real part of ε already becomes negative for $n = 0.1$, whereas for zero electric field it take place only for $n \sim 0.5$.

All previous results were obtained for the strong electric field, when Stark shifts essentially exceed not only the natural width γ of excited states of the free atom, but also the typical level shifts caused by dipole-dipole interatomic interaction. Figure 4 demonstrates that noticeable effects of this field are important even for a Stark shift comparable with γ, i.e., for $\Delta \sim 1$. The curves in Fig. 4 were calculated for an atomic ensemble with $n = 0.15$.

Figure 4 shows the qualitative difference in the influence of a relatively small electric field on the spectrum of the absorbtion line for two principal polarizations. For circularly polarized coherent light [Fig. 4(a)], this field causes a noticeable shift but small modification of the maximum of absorption. For the linear polarization [Fig. 4(b)], on the contrary, the shift is small and the amplitude is essentially increased. As the strength of the static field increases, the absolute value of k'' as well as the shift of the spectral line increases for both cases. For $\Delta > 10$, the dependence of k'' on Δ practically disappears. For such a field, the Stark shifts essentially exceed the level shifts caused by interatomic interaction.

IV. CONCLUSION

In the present work, we study the influence of an electrostatic field on the optical properties of dense and cold atomic gases prepared in a special atomic trap. We calculate the dispersion of the permittivity tensor for different field strengths and for different atomic densities. We show that the manifestation and the mechanism of electro-optical effects in this case differs essentially from the case of dilute atomic ensembles.

For dense gases, the influence of an electrostatic field does not restrict itself to a Stark shift of optical resonances. We observe here the alteration in the width, amplitude, and shape of atomic transitions. The electric field suppresses the saturation effect which takes place for dense gases [26,40]. In an electric field, the resonances become narrower and their amplitude increases. The electric field also decreases the collective Lamb shift. It opens additional possibilities to control over the optical properties of dense and cold atomic ensembles.

Our analysis reveals that the main reason for the modification of optical properties of the dense gases in an electric field is the modification of polyatomic collective effects caused by interatomic dipole-dipole resonant interaction. In the considered case, such an indirect influence is much more effective than direct transformation of optical properties caused by single-atom effects in an electrostatic field.

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