Nonlinear electro-optic effects in resonant atomic media: a novel tool for precision atomic spectroscopy and electromagnetic field measurements

(A Proposal)

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Nonlinear electro-optic effects in resonant atomic media: a novel tool for precision atomic spectroscopy and electromagnetic field measurements

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We propose to apply nonlinear electro-optic effects in atoms and molecules to sensitive measurements of fundamental atomic and molecular parameters (e.g., tensor polarizabilities, permanent dipole moments). We also propose the use of these effects for non-intrusive, spatially and temporally resolved electromagnetic field measurements (electromagnetic field tomography).

I. INTRODUCTION

We propose to study nonlinear optical effects which arise when near-resonant light propagates in an atomic medium in the presence of an electric field. We will utilize techniques similar to those we have used in the study and application of nonlinear magneto-optic effects (NMOE) [1,2]. The nonlinear electro-optic effects (NEOE) and NMOE display both similarities and essential differences as will be discussed below, but both can be used to achieve sensitivity to minute energy level shifts (down to $\sim 10^{-6}$ Hz/$\sqrt{\text{Hz}}$; see [1–3]).

In recent years, considerable amount of work has been devoted to the investigation and applications of NMOE; see e.g. [4,5] and references therein. These effects lead to drastic modification of optical rotation (the rotation of light polarization plane) in the presence of a magnetic field compared to the linear case. In particular, by employing long-lived atomic coherences the magnitude of the rotation can be enhanced by up to nine orders of magnitude [1].[1] This enhancement makes NMOE a powerful tool for magnetic field measurements [12,2,3], with shot-noise limited sensitivities of $\sim 10^{-12}$ G/$\sqrt{\text{Hz}}$ [3], surpassing that of the best optical pumping and SQUID magnetometers.

NEOE could have applications both for probing fundamental properties of atoms and for precise electric field measurements. NEOE may allow electric field measurements in situations where other methods are ineffective. For example, this method would be non-intrusive: except for the minute amounts of atoms present in the volume where the electric field is measured (corresponding to high vacuum conditions), there is no electric field "probe" that needs to be introduced.

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[1] Long-lived atomic coherences are also responsible for a wide range of spectacular nonlinear optics phenomena [6] that have been investigated in the last decade and applied in a variety of technologies, including subrecoil laser cooling [7], electromagnetically-induced transparency [8], and slow group velocity of light [9–11] (the slowest velocity 8 m/s was achieved in our work [11]).

Below, we briefly review earlier work on NEOE, present calculations of NEOE with simple atomic systems in the absence of atomic collisions, and estimate the sensitivity of NEOE measurements. We further discuss possible applications of NEOE and the details of the proposed research, including both theoretical and experimental studies.

II. EARLIER WORK ON NEOE

Although the linear electro-optic effect in resonance media has been known and used for Stark shift measurements since the 1920's (see [13] for a comprehensive review), up to now there has been relatively little work on NEOE. An important exception is the work of the Oxford group [14]. Using the modulation polarimetry method, they investigated linear and nonlinear light polarization rotation in the vicinity of $J = 0 \rightarrow J' = 1$ transitions in atomic samarium. They observed dramatic differences in the spectrum and magnitude of the rotation between the two cases, provided qualitative explanations for their observations, and developed a density matrix theory which gave a satisfactory quantitative description. The complexity of the theoretical calculations was minimized due to the low electronic angular momenta involved and through the use of a number of simplifying approximations that may not be applicable in many situations (particularly, for the case of alkali atoms with complicated hyperfine structure and in the presence of collisions). Further theoretical work on NEOE was done by Fomichev [15] who considered $0 \rightarrow 1$ and $1 \rightarrow 0$ transitions in the presence of an arbitrarily directed DC electric field.

In work closely related to NEOE, the Amherst group [16] investigated modification of the optical properties of optically pumped atoms (Ba and Cs) in the presence of crossed electric and magnetic fields. This research was carried out in conjunction with experiments searching for the parity- and time reversal invariance violating electric dipole moment (EDM) of the electron [17]. The technique used by this group to search for the EDM, involving optical detection of atomic angular momentum precession in an electric field, can also be thought as a variety of NEOE. Similar techniques are also used to search for EDM of diamagnetic atoms (Hg, Xe; see e.g. [18] and references therein).

III. NEOE IN SIMPLE SITUATIONS

Consider a collision-free gas of atoms with zero nuclear spin in a $J = 1$ ground state. Suppose the atoms are illuminated with monochromatic light propagating in the $\hat{x}$ direction which is linearly polarized along $\hat{y} + \hat{z}$ (Fig. 1). The light frequency is tuned near resonance with a transition to a state with $J' = 0$. A DC electric field is applied along $\hat{z}$.

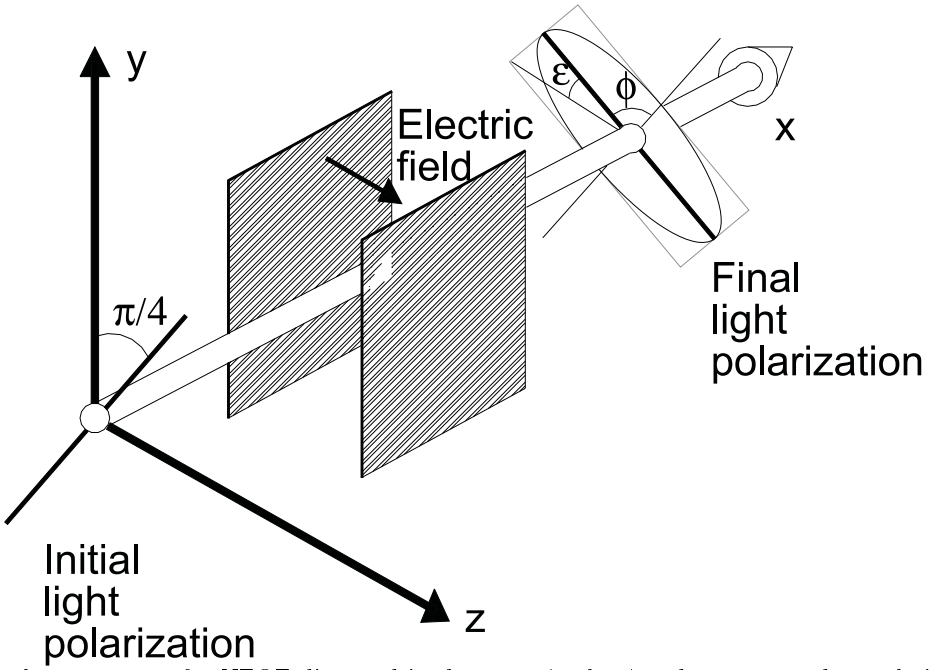

FIG. 1. Geometrical arrangement for NEOE discussed in the text. Angles ϕ and ϵ correspond to polarization rotation and ellipticity, respectively.

Due to tensor polarizability of the ground state, the $m = \pm 1$ and $m = 0$ Zeeman components are split in energy in the presence of the electric field. This differential shift leads to a difference in resonance frequencies for the linear light polarization components along $\hat{z}$ and $\hat{y}$. Thus, the medium becomes optically anisotropic, in general exhibiting both birefringence (difference in refraction) and dichroism (difference in absorption). Below, we present the results of a theoretical analysis of these effects (based on density matrix calculations) at low (non-saturating) and high (saturating) light intensity, producing linear and nonlinear effects, respectively.

We begin the analysis with the case of the linear electro-optic effect for a closed optical transition. Fig. 2 a,b shows the rotation ϕ and ellipticity ϵ as a function of light frequency detuning from the center of the field-free resonance. The rotation and ellipticity are given per unsaturated absorption length (on resonance), and the value of the Stark shift is chosen to be $\Delta_S = \gamma/2$, where $\gamma = 2\pi{\cdot}6$ MHz is the natural width of the transition (i.e. the shift is a small fraction of the Doppler width, $\Gamma_D = 2\pi{\cdot}300$ MHz; we set $\hbar = 1$ throughout). The light intensity is much less than the saturation intensity for the transition. The spectral dependences and magnitudes of ϕ and ϵ are consistent with what one expects from linear optics. Note that the maximum values of ϕ and ϵ in the linear case are approximately equal to each other (Fig. 2 a,b).

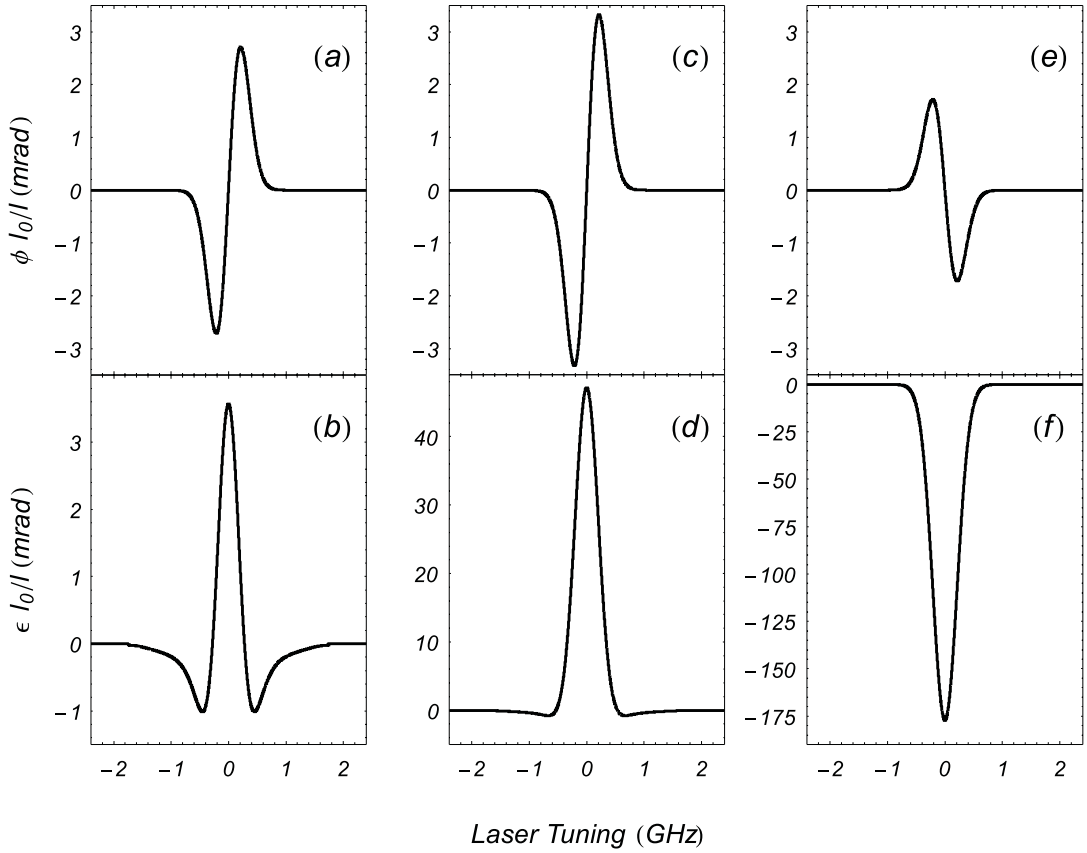

FIG. 2. Optical rotation ϕ and ellipticity ϵ per unsaturated absorption length (at the center of the field-free resonance) as a function of light frequency detuning from the center of the field-free resonance. a, b: linear effect; c, d: laser power is increased so ϕ and ϵ are primarily due to the Bennett structure effect; e, f: DC electric field is reduced so ϕ and ϵ are primarily due to the coherence effect. The characteristic width of all these curves is determined by the Doppler width $\Gamma_D/2\pi$=300 MHz. Various other parameters used in the calculation are discussed in the text. With these parameters, the magnitude of the rotation is approximately the same for the three plots, whereas ellipticity differs by over an order of magnitude.

Next, we increase the light intensity to the saturation intensity, at which absorption per unit length is decreased by a factor of 2 with respect to low power absorption. With effective relaxation of the ground state populations $\gamma_{rel} = 2\pi \cdot 0.06$ MHz due to the transit of atoms through the laser beam and the chosen value of the transition dipole moment $d = 1e \cdot a_0$, the saturation intensity is approximately 0.33 mW/cm^2. The rotation and ellipticity in this case are shown in Fig. 2 c and d, respectively. We see that while there is relatively little change in rotation compared to the linear case (see Fig. 2 a,c), there are significant differences in the magnitude and the spectral line shape for the ellipticity (Fig. 2 b,d). This nonlinear effect in ellipticity is related to the light-induced Bennett structures in the velocity distribution of the ground state atoms (a similar effect is observed in magneto-optical rotation, see e.g. [5,19] and references therein). Qualitatively the electric field-induced ellipticity is due to the difference of the dispersively-shaped real parts of the refractive index. Due to the narrow ($\approx \gamma$) Bennett structures, the maximum ellipticity becomes proportional to Δ_S/γ, rather than Δ_S/Γ_D as in the linear case. There is no such enhancement for

optical rotation ϕ because the rotation depends on the difference in the bell-shaped imaginary parts of the refractive index. This difference is zero for the resonant velocity group, and of opposite signs for the velocity groups to either side of the resonance. Therefore, the overall effect is only nonzero due to the finite Doppler width.

If we now reduce the DC electric field, so that $\Delta_S = \gamma_{rel}/2$, leaving all other parameters unchanged, we see that despite the fact that the Stark shift was decreased by a factor of a hundred, the ellipticity is about three times larger than in the Bennett structure effect, and that both ellipticity and rotation have reversed sign (Fig. 2 e,f). This is a coherence effect, similar to those found in NMOE, with maximum ellipticity of magnitude proportional to Δ_S/γ_{rel} (for $\Delta_S \ll \gamma_{rel}$). The mechanism for a coherence effect can be thought of as a three-step process (also discussed in the context of NMOE in [20,1,5]). In the first step, linearly polarized light produces atomic alignment along the polarization direction. In the second step, this alignment evolves in the electric field. In the magnetic field case, the evolution corresponds to precession of the alignment around the direction of the field. For an electric field, the evolution is of a more complicated character [21], involving oscillation of the alignment and appearance of other oscillating moments (orientation, and higher rank moments for $J \geq 1$). An example of the evolution of atomic alignment corresponding to our example of $J = 1$ is shown in Fig. 3. The third step consists in the interaction of the light with an atomic system with the evolved polarization. This interaction modifies the polarization of the transmitted light.

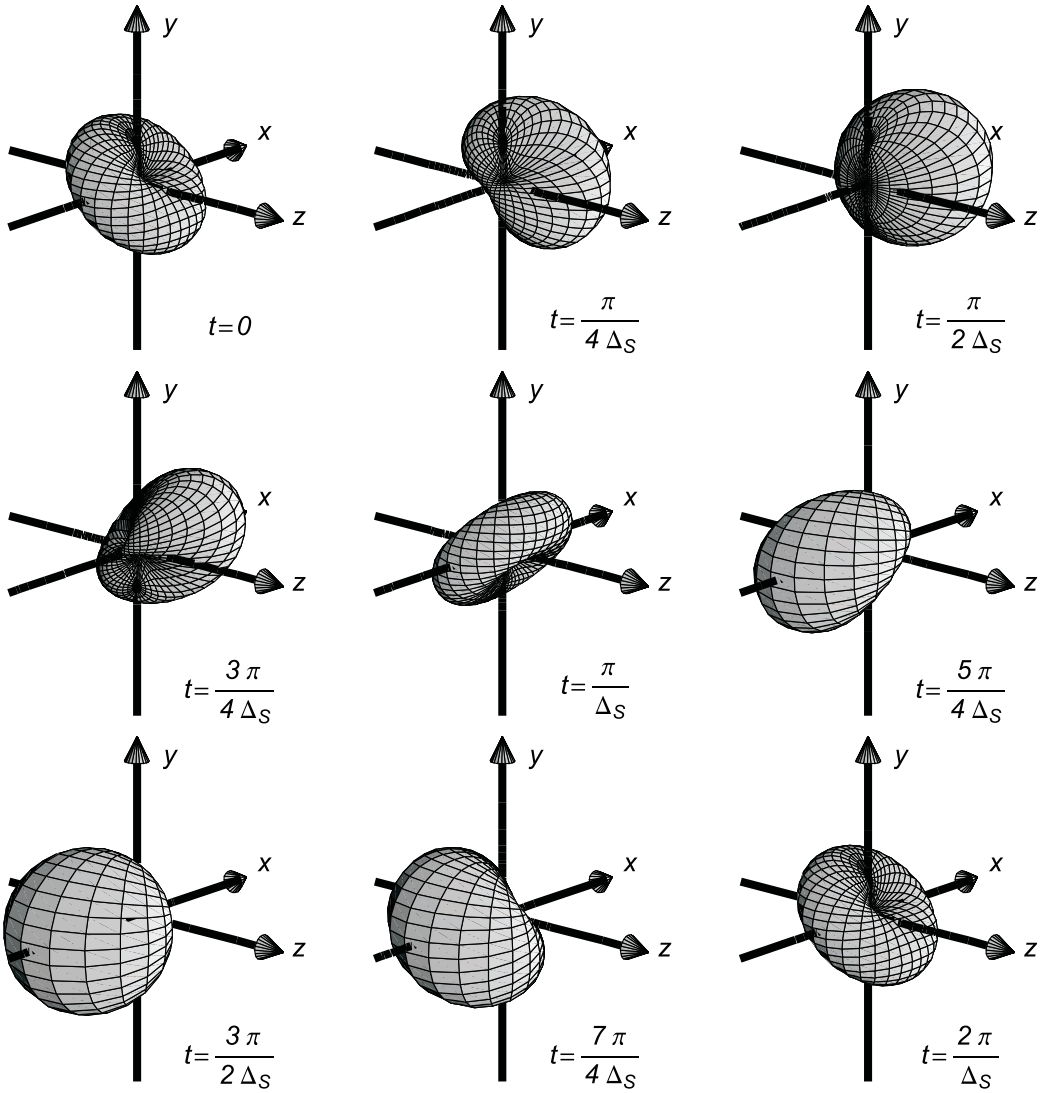

FIG. 3. Temporal evolution of a $J = 1$ system prepared at $t = 0$ in a state aligned along $\hat{z} + \hat{y}$ (the $m = 0$ state for the quantization axis along $\hat{z} + \hat{y}$) in an electric field along $\hat{z}$. Each of the plots shows a surface whose distance from the origin at a given direction is proportional to the probability of finding $m = 1$ in that direction [21].

The various NEOE effects (linear, Bennett structure, and coherence) can be seen on the plots of ϵ (Fig. 4) as a function of the Stark shift Δ_S with the light at saturation intensity and tuned to the field-free resonance. These plots are similar to those for ϕ vs. Zeeman shift used to distinguish the corresponding effects in NMOE [12,1,2]. Note that the ellipticity induced by small Stark shifts is greater by many orders of magnitude in the case of NEOE compared with the linear effect.

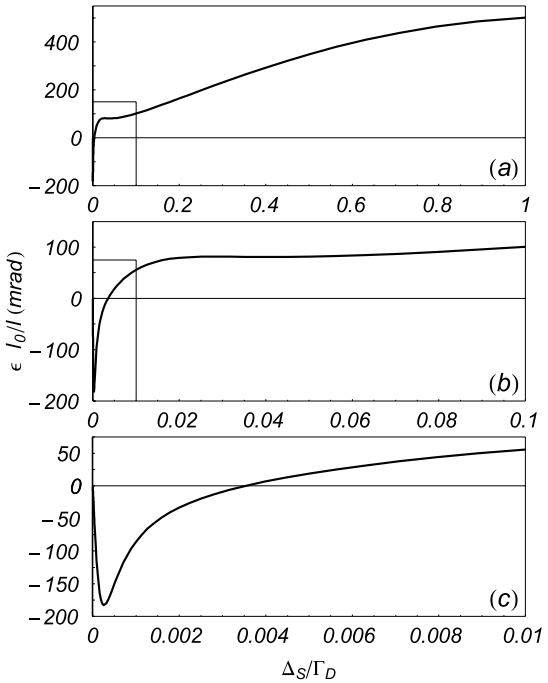

FIG. 4. Ellipticity per unsaturated absorption length as a function of the Stark shift normalized to the Doppler width. *a*: linear effect; *b*: Bennett structure effect; *c*: coherence effect. The light frequency is tuned to the electric field-free resonance. Various parameters used in the calculation are discussed in the text.

For alkali atoms in paraffin coated cells γ_{rel} can be made $\leq 2\pi \cdot 1$ Hz [22]. The paraffin coating allows thousands of wall collisions without depolarization. Atoms aligned by the laser light wander around the vapor cell, eventually returning back to the laser beam and interacting with it again. This leads to the ultra-narrow features in the field dependence of NMOE and NEOE known as the "wall-induced Ramsey effect." However, it is important to realize that because alkali atoms have electronic angular momentum $J = 1/2$ in the ground state, their tensor polarizabilities are suppressed by $\sim \omega_0/\Delta_{hfs} \sim 10^5$ with respect to the scalar polarizabilities, where ω_0 is the energy of the lowest P-states and Δ_{hfs} is a typical ground state hyperfine splitting. Therefore, the use of paraffin-coated cells in applications relying on the ground state tensor polarizability may not be practical.

Recent experimental evidence indicates a possibility of obtaining extremely low values of γ_{rel} (≤ 0.03 Hz) for alkali atoms in buffer gas cells at $T \approx 2$ K [23]. At room temperature, the smallest values of γ_{rel} that can be achieved in buffer gas cells are on the order of $20 - 50$ Hz [17,24]. However, at $T \approx 2$ K, atomic collisions become predominantly S-wave, which leads to a significant reduction of the depolarization cross sections [25]. Techniques for atomic spectroscopy at these temperatures are well-developed and the necessary cryogenic equipment is available commercially (a research cryostat with optical access can be purchased for $\sim$ \$10 k). Experimental investigation of the nonlinear magneto- and electro-optic effects with such narrow widths appears both feasible and promising for ultra-precise atomic spectroscopy and fundamental measurements.

IV. APPLICATIONS OF NEOE

1. Non-intrusive electric field measurements. Electromagnetic field tomography. It is a common problem both in research and technological applications (including aerospace) to measure the electric field strength in a certain location without perturbing this field or causing electrical breakdown. In many cases, the fields need to be measured either in vacuum or in the presence of a gas of a well-defined composition in the test volume. These requirements may be related, for example, to the necessity of maintaining high electrical breakdown threshold. We propose to use NEOE for such non-intrusive measurements.

A schematic of the method is shown in Fig. 5. To make the measurement, a trace amount of atoms (corresponding to a vapor pressure $\sim 10^{-7}$ Torr) is introduced into the system. For example, if a solid rubidium particulate is introduced into a test volume at room temperature, an equilibrium vapor pressure of $\approx 3{\cdot}10^{-7}$ Torr will be established throughout the volume. Other means of introduction of atoms or molecules into the test volume could be used, e.g. atomic or molecular beams. A pump laser beam intense enough to induce the nonlinear effects in the "probe" atoms is directed through the test volume. The initial polarization of the pump laser beam is set (e.g. to a linear polarization) by polarizing elements. The polarization state after propagation through the test volume then contains information about the fields in the volume contained inside the pump beam. However, to further localize the measurement area, a weak probe laser beam (with intensity low enough so that there are no nonlinear effects) is intersected with the pump beam. (This beam could be derived from the same laser as the probe beam.) The polarization state of the probe beam is prepared appropriately by polarizing elements and the polarization state after propagation through the test volume is analyzed by a polarization analyzer (e.g. a circular analyzer). The laser, the polarization elements and the analyzer could all be positioned in convenient locations, for example, outside the test volume and arbitrarily far from it. By detecting the probe output polarization state, it is possible to reconstruct the magnitudes and directions of both the magnetic and electric fields in the region of the intersection of the pump and probe beams. This reconstruction is aided by the fact that NMOE involve enhanced polarization rotation, while NEOE feature enhanced ellipiticity, allowing separation of the electric and magnetic field effects. Scanning the laser beam intersection position, one can obtain three-dimensional information about electric and magnetic fields in the test volume (electromagnetic field tomography). In addition, one could consider working with large laser beams and having a detector with spatial resolution (e.g., a CCD camera). This may allow simultaneous measurement of electromagnetic fields in an array of points and without spatial scanning of the laser beam [26]. Note also that the high statistical sensitivity of the

method allows precise measurements to be made quickly, thus enabling measurement of time-dependent fields.

The sensitivity of the method to electric fields depends on the concrete problem and details of the realization. As an example, let us consider electric field measurements in our recent experiment on lifetimes and quantum beats in Sm [27]. There, it was necessary to precisely measure electric fields ~ 20 kV/cm. Suppose that one uses Sm atoms (already available in that experiment) in the $J = 1$ ground term state and the coherence NEOE with transit relaxation with $\gamma_{rel} \sim 2\pi \cdot 100$ kHz. Using the ground state tensor polarizability of α_2=0.28 kHz/(kV/cm)^2 [28], the tensor Stark shift is $\Delta_S \sim$ 100 kHz. Thus if the probe region comprises ~ 1 absorption length, the ellipticity acquired by the probe light is on the order of 0.2 radian (see Fig. 2 f). With a milliwatt laser, the ellipticity can be measured with statistical sensitivity $\sim 10^{-8}$ rad/$\sqrt{Hz}$. This translates into a resolution of $\delta E/E \sim 10^{-7}$ or $\delta E \sim 2$ mV/cm. Note that an apparatus for these measurements is already essentially in place in our laboratory [27].

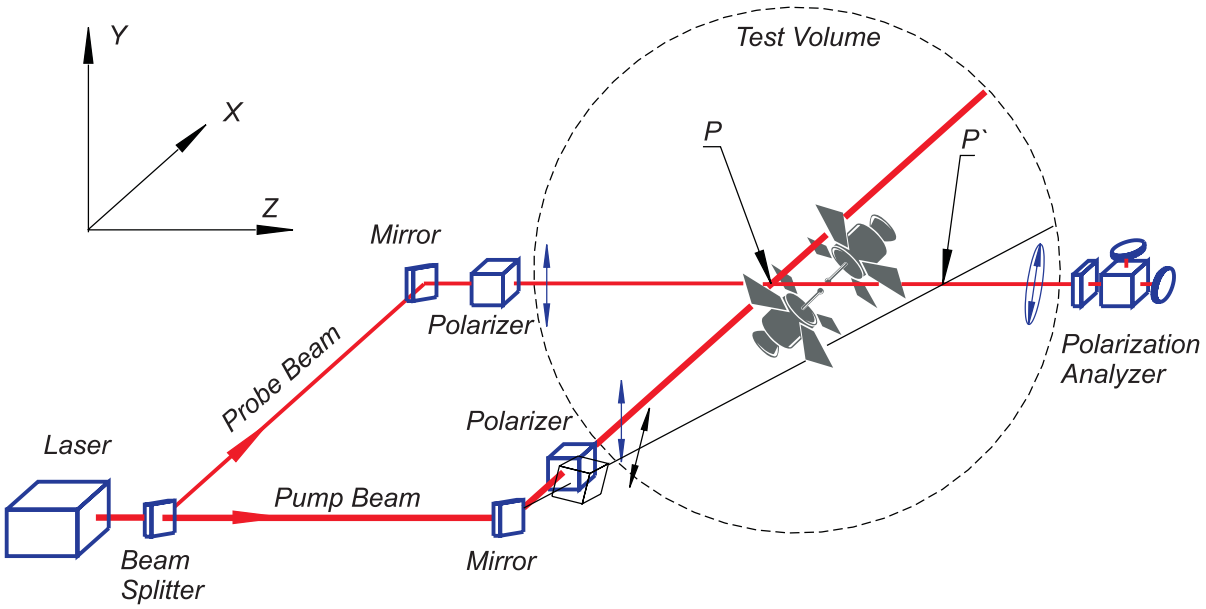

FIG. 5. Schematic for electromagnetic field measurement.

2. Determination of atomic parameters.

NEOE can be applied to measurements of atomic parameters, e.g. tensor electric polarizabilities, in two ways: directly (if the electric field is known) and as a method of electric field calibration. The direct method was applied to measurements in samarium [14]. It appears that both direct and indirect application of NEOE could improve the precision of the tensor polarizability measurements in alkali atoms (see e.g. [29] and references therein). These measurements have currently reached the level of experimental uncertainties of $\sim 2 \cdot 10^{-3}$ and are important as checks for ab-initio and semi-empirical atomic calculations that are employed in the work on frequency standards, atomic

parity violation, etc.

3. Search for atomic EDM. If the electric field is applied along the direction light propagation, rather than perpendicular to it, optical rotation violates parity and time-reversal symmetry. Application of NEOE in the search for P- and T- violating permanent electric dipole moment was first suggested in [12], and later discussed in [30,17]. The sketch of a possible EDM experiment is, in general, very similar to the one used to observe the NMOE dependence on B_z [2]. The only principal difference consists in the detection of alignment precession caused by interaction of atom's P- and T- violating EDM d_A with applied electric field E. The shot-noise-limited sensitivity δd_A to the measurement of EDM may be found from the smallest detectable change of the magnetic field, δB_z, according to the relation:

$$\delta d_A = \frac{g_J \mu}{E} \delta B_z, \tag{1}$$

where $g_J = 2$. With the sensitivity $\delta B_z \sim 10^{-12}$ G$/\sqrt{\text{Hz}}$ estimated in our NMOE work with rubidium in coated vapor cells [3], one obtains:

$$\delta d_A \approx 10^{-27}\ e \cdot \text{cm}, \tag{2}$$

assuming electric field $E = 10$ kV/cm and data accumulation time $T = 10^6$ sec. This corresponds to a sensitivity to the electron's EDM, d_e that is, according to theory (see, e.g. [31] and references therein) approximately 120 times smaller:

$$\delta d_e \approx 10^{-29}\ e \cdot \text{cm}. \tag{3}$$

To obtain an EDM limit at the level of statistical sensitivity (2), one has to control possible systematic effects at least at a similar level. One method of doing this is to conduct an experiment with a coated cell containing vapors of both Cs and Rb atoms [32]. The EDM measurements are performed on Cs atoms, and Rb is used as a "co-magnetometer." NEOE can also be used to precisely measure the applied electric field, as described above.

The limits (2,3) are more than three orders of magnitude better than the current limit for d_{Cs} [33] and about 400 times better than the best published experimental limit on electron EDM, $|d_e| \leq 4 \cdot 10^{-27}\ e$·cm, established in an experiment with ^{205}Tl [34].

V. THE PROPOSED RESEARCH

We propose to carry out the following research:

– Experimentally investigate application of electric fields in coated cells. We will use NMOE and NEOE to measure the magnitude, uniformity and stability of the electric fields in such systems and possible magnetic field distortions due to leakage currents. The effects of the spin-exchange collisions between rubidium and cesium in the cell will be explored. We will use the results of this research to evaluate the prospects for precision EDM and tensor polarizability measurements using NEOE.

– Explore NEOE in cold (~ 2 K) buffer gas cells. Investigate the possibility of atomic energy shift measurements of unprecedented sensitivity.

– Design and implement a prototype device for three-dimensional electromagnetic tomography. One of the first applications of this device could be a study of the so-called patch and surface charge effects that are known to be a limiting factor in high-voltage systems where precise magnitudes and reversals of the electric fields are necessary (e.g. for tensor polarizability and EDM measurements).

– Extend the calculations of NEOE presented above to multi-level alkali atoms which are important for many practical applications. Develop a complete understanding of these complex systems and create efficient software for detailed quantitative calculations allowing extraction of the electric field parameters from the experimental observations.

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