INSTRUMENTS AND METHODS OF INVESTIGATION

On experiments in search of the T-odd electric dipole moment of atomic ¹²⁹Xe

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<u>Abstract.</u> A new method is proposed for the search of the electric dipole moment of atomic ¹²⁹Xe arising from time reversal symmetry violation. We consider the basic features of the experiment to measure the electric field of T-odd EDMs of spin-polarized ¹²⁹Xe atoms with the aid of a Stark electrometer involving Rydberg atoms.

1. Introduction

The preamble of our paper is dedicated to a brief review of experiments in search of the electric dipole moment (EDM) in quantum systems and the current status of the problem. For more details of the EDM of quantum systems, see Refs [1, 2].

1.1 On the electric dipole moment of quantum systems

An isolated quantum system in a nondegenerate stationary state may have a permanent EDM under violation of the symmetry with respect to the time reversal operation T. The CP symmetry must be simultaneously violated, where C is the operation of charge conjugation and P is the inversion of spatial coordinates. This statement follows from the CPTinvariance requirement.

From the very outset, there is good reason to mention that the obvious picture of an HCl-like molecule, in which the electric dipole moment is aligned with the molecular axis, is irrelevant to the problem under discussion. This picture corresponds to a molecule retained by external forces, i.e., is

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Received 31 January 2005, revised 16 February 2005 Uspekhi Fizicheskikh Nauk **175** (9) 979–993 (2005) Translated by E N Ragozin; edited by A M Semikhatov not isolated and cannot be assigned a specific stationary state. The latter is characterized by the quantum numbers of the total angular momentum $J^2 = J(J+1)$ and its projection $J_z = m$ on an arbitrarily selected axis in space. In the state J, m, the electric dipole moment is equal to zero. It is averaged owing to molecular rotation.

We therefore consider an isolated system in a nondegenerate stationary state — elementary particles, an atomic nucleus, an atom. In the frame of reference in which the system is at rest, it may be characterized by a single vector intrinsic angular momentum — the spin J. The system's EDM **d** can only be directed along J,

$$\mathbf{d} = A\mathbf{J}, \qquad d_z = AJ_z. \tag{1.1}$$

Under spatial coordinate inversion P, the polar vector **d** changes sign and the axial vector **J** does not. Under time reversal T, the vector **d** does not change and the vector **J** changes sign. This implies that the constant A in expression (1.1) can take only one value: A = 0. Therefore, Eqn (1.1) implies that the EDM may be nonzero only on the strength of the violation of both the P and T symmetry.

After the P-symmetry violation in electroweak interactions was demonstrated in three almost simultaneous classic experiments in 1956–1957 (see Refs [1, 2]), in 1957 Landau already showed [3] that for the EDM to exist in elementary particles, the violation of weak-interaction symmetry with respect to the P and C operations taken separately would not suffice and that the CP-invariance violation is required: "It is easy to show that particles cannot possess dipole moments in the scheme outlined, despite the absence of conventional parity." From the CPT invariance, it follows that the EDM is simultaneously prohibited by the CP symmetry and by the T symmetry. Therefore, the T- and CP-symmetry violation problems turned out to be interdependent.

In that work, Landau also formulated a statement of significance to the EDM problem, which was invoked in Eqn (1.1): the only vector that can be formed for a particle at

rest is the vector of its spin, which is inversion-even and charge-odd.

As a result, the emphasis in the discussion of the EDM problem has been on its relation to T-symmetry violation, i.e., on time reversal symmetry. At about the same time, approximately 50 years ago, a start was made on experiments in search for the neutron EDM.

1.2 Topicality of the EDM problem

The CP-symmetry violation in the decay of K⁰ mesons was discovered in 1964. In 2001, the CP-symmetry violation was also discovered in the decay of B^0 mesons. The first experimental results on the decay of K mesons bolstered interest in experiments in search for an EDM in different quantum objects, primarily in experiments searching for the neutron EDM, already underway. It was not long before a start was made on the search for the EDMs of electrons, atoms, and molecules [1, 2, 4]. The upper limits were established for the EDM of neutrons and electrons, as well as some paramagnetic and diamagnetic atoms. These findings proved to be highly important to elementary particle physics because they permitted formulating several limitations on the theoretical models of CP-symmetry violation. The most important part was played by the ascertainment of the upper bound for the neutron EDM.

The presently accepted upper bound for the neutron EDM [1, 2]

$$d_{\rm n} \leqslant 6.3 \times 10^{-26} e \,\,{\rm cm}\,,\tag{1.2}$$

where *e* is the electron charge, is the outcome of experimental work approximately 50 years long. Many theories proposed to account for the CP-symmetry violation have been rejected because they predict neutron EDM values above the experimental upper bound in (1.2).

The standard model, which is presently the most popular in elementary particle physics and nuclear physics, predicts substantially lower neutron EDM values at about 10^{-32} – $10^{-34} e$ cm. For the EDM of other quantum objects, the standard model also predicts very small values, which do not lend themselves to evaluation by contemporary experimental techniques. However, several extensions of the standard model predict significantly higher EDM values than the standard model itself (see, e.g., Ref. [2]). As a consequence, interest in new experiments to search for the EDM of quantum objects has substantially grown because it was recognized that a new physics that does not fall under the province of the standard model is extremely sensitive to the results of these experiments. Khriplovich [5] summarizes the situation as follows: "As to the mechanism of CP violation incorporated in the standard model of electroweak interactions, which is the most popular at present, the prediction for the neutron EDM is six orders of magnitude below the present experimental bound. The gap for the electron EDM is much larger.

"But does this mean that EDM experiments are of no serious interest for elementary particle physics, that they are nothing but mere exercises in precision spectroscopy? Just the opposite. It means that these experiments now, at the present level of accuracy, are extremely sensitive to a possible new physics beyond the standard model, a physics to which the kaon decays are insensitive?"

Hopes have risen that the necessary new experiments may be conducted even on the basis of contemporary

experimental technique, upon its complex and yet realistic improvement. For more details on the EDM of quantum systems, see Refs [1, 2].

1.3 Experiments in search of the neutron EDM

Among the experimental investigations in search of the EDM, the neutron is in the lead. The grounds for this are discussed in Refs [1, 2] as well as in the program "A new precision measurement of the neutron electric dipole moment" [6]. Ten international centers participate in this program, three of which are located in Russia: the Ioffe Physicotechnical Institute of the Russian Academy of Sciences, the Konstantinov Institute of Nuclear Physics of the Russian Academy of Sciences, and the Vavilov State Optical Institute. The program has a highly ambitious goal: starting from the d_n value in (1.2), to reach the level $d_{\rm n} \sim 2 \times 10^{-28} \, e \, {\rm cm}$ by 2010.

The following basic idea underlies the experiment. In collinear magnetic **B** and electric **E** fields, the spin of a particle that has a magnetic moment μ and an electric dipole moment **d** precesses with the frequency

$$\Omega_{\pm} = \frac{2|\mu B \pm dE|}{\hbar} \,, \tag{1.3}$$

where the + sign corresponds to parallel and the - sign to antiparallel B and E. The factor 2 in (1.3) arises because the amplitudes of μ and d (of magnetic and electric dipole moments, respectively) are defined as $\mu = \mu \sigma$ and $\mathbf{d} = d\sigma$, where σ is the Pauli spin operator. On reversing the direction of the field E, the precession frequency must change by the value

$$\Delta \Omega = \frac{4dE}{\hbar} \,. \tag{1.4}$$

The EDM d is found from the experimental value of $\Delta \Omega$ using formula (1.4). In the realization of this procedure, there emerges an uncertainty arising from the necessity of extracting the sought effect $\Delta \Omega$ against the background of the precession frequency fluctuations $\delta \Omega$ of a statistical nature $(\delta \Omega_{st})$, as well as fluctuations due to numerous spurious instrumental effects ($\delta \Omega_{instr}$).

The quantity μB in (1.3) exceeds the quantity dE by many orders of magnitude, and therefore the main contribution to $\delta\Omega$ is made by magnetic field instability, arising for several reasons, including its variation due to leakage currents correlated with the change of the electric field direction, misalignment of the fields **B** and **E**, etc.

The quantities $\delta \Omega_{st}$ and $\delta \Omega_{instr}$ essentially depend on the specific experimental setup and technique, as well as on the characteristics of the experimental facility.

The program [6] involves the following research phases:

Phase I (2004–2005) $\rightarrow d_n \sim 10^{-26} e$ cm, Phase II (2006–2007) $\rightarrow d_n \sim 10^{-27} e$ cm,

Phase III (2008–2010) $\rightarrow d_{\rm n} \sim 2 \times 10^{-28} e$ cm.

It is planned that the special-purpose experimental complexes available for program participants will be substantially updated and entirely new complexes will be made. A significant intensity increase of the source of ultracold spinpolarized neutrons (UCNs) is projected. A new EDM spectrometer reliant on the resonance Ramsey technique is contemplated for installation. The spectrometer will have four working storage volumes for UCNs with twin chambers, with the imposed electric field E parallel to the magnetic field **B** in one of them and antiparallel to **B** in the other. In five additional chambers, the electric field is absent. Sixteen Cs magnetometers will exert control over the magnetic field. The electric field-free chambers should furnish additional information on the magnetic field, fulfilling the function of neutron magnetometers. All the 13 storage chambers and 16 Cs magnetometers are arranged in such a way as to ensure the requisite control and suppression of spurious systematic instrumental effects by way of combined processing of the measurements of 13 independent shifts in the precession frequency Ω . During the concluding phase of the work, it is contemplated to ensure the following main parameters of the experimental facility.

• Filling of the four working chambers with ultracold spin-polarized neutrons with the periodicity $\sim 10^3$ s.

• The kinetic neutron energy

$$\varepsilon_{\rm k} \approx 2.5 \times 10^{-7} \text{ eV} \,. \tag{1.5}$$

• The neutron density in the working chamber

$$N = 3 \times 10^3 \text{ UCN cm}^{-3}.$$
(1.6)

• The total volume of the working chambers and the total number of neutrons

$$V = 2 \times 10^5 \text{ cm}^3$$
, $N_0 = NV = 6 \times 10^8 \text{ UCN}$. (1.7)

• The magnetic and electric intensities

$$B = 2 \times 10^{-6} \text{ T}, \quad E = 15 \times 10^3 \text{ V cm}^{-1}.$$
 (1.8)

• The precession frequency Ω in (1.3) and the quantity $\Delta \Omega$ in (1.4) corresponding to the *d* values stated in the program should have the values

$$\frac{\Omega}{2\pi} \sim 5 \times 10^2 \text{ Hz}, \quad \frac{\Delta\Omega}{2\pi} \sim 2 \times 10^{-8} \text{ Hz}, \quad \frac{\Delta\Omega}{\Omega} \sim 4 \times 10^{-11}.$$
(1.9)

A comprehensive review of all possible systematic spurious effects and ways to control them was made by Aleksandrov et al. [6].

Of special significance to the subsequent discussion is the fact that the value of the main quantity to be measured,

$$\frac{\Delta\Omega}{2\pi} \sim 2 \times 10^{-8} \text{ Hz}, \qquad (1.10)$$

is a small addition (~ 4×10^{-11}) to the precession frequency Ω , which emerges due to the existence of the neutron EDM $d_n \sim 10^{-28} e$ cm.

The experimental data acquisition time required to determine the neutron EDM to within $2 \times 10^{-28} e$ cm is estimated at T = 1 yr [6] (assuming the fraction of 'pure' experimental time to be equal to 30%).

1.4 Atomic EDM. Schiff's theorem

The discussion of the EDM problem of a neutral atom brings up the question: What EDM can an atom have if its nucleus or electrons have an EDM? This problem was discussed in detail in Ref. [1]. The answer can be briefly formulated as follows. Assuming point charges coupled by electrostatic interaction, an atom cannot have an EDM d_A , even though the atomic nucleus and the electrons may have intrinsic EDMs, d_N and d_e .

The aforesaid is a consequence of a peculiar screening principle, which is quite frequently referred to as Schiff's theorem. This theorem applies to any system of point particles that are retained in a bound equilibrium state by electrostatic interaction. The presence of EDM in the particles of the system results in their configuration redistribution, in comparison with the configuration for zero EDMs, such that the total EDM of the system is equal to zero. In particular, the nucleus EDM and the electron EDMs are screened such that no EDM is induced in the atom.

The prohibition imposed on the existence of an EDM in a system due to the screening effect is violated in two cases: if finite particle dimensions are taken into account or if the magnetic interaction is included along with the electrostatic one. Schiff obtained this result in his fundamental work [7] concerned with the EDM of the nucleus of atomic ³He. The prohibition of the EDM for neutral atoms may also be removed for similar reasons, which has allowed the performance of experiments in search of atomic EDMs.

1.5 Paramagnetic atoms

For paramagnetic atoms, of primary interest is the atomic EDM caused by the electron EDM d_e .

Sandars [8] showed that the Schiff theorem breaks down for paramagnetic atoms due to magnetic interactions, an important 'enhancement' effect taking place for the electron EDM. The ratio $R = d_A/d_e$ is of the order of $10Z^3\alpha^2$, where Z is the atomic nuclear charge and $\alpha = 1/137$ is the fine structure constant.

The values of R calculated for several atoms are given in Table 1 (see Refs [1, 2]).

The most significant bound on the electron EDM was obtained in Berkley [9]. The final result, published in 1994 [9], is

$$d(^{205}\text{Tl}) = (-1.05 \pm 0.70 \pm 0.59) \times 10^{-24} e \text{ cm}.$$
 (1.11)

Assuming that there is no other contribution to the EDM of atomic Tl apart from the electron EDM and adopting the 'enhancement' factor R from Table 1, it is possible to obtain the upper bound for the electron EDM. A paper published in 2002 gives the value [9]

$$d_{\rm e} = (6.9 \pm 7.4) \times 10^{-28} \, e \, {\rm cm} \,. \tag{1.12}$$

Table 1.

Atom	Ζ	Ground state	R	
Na	11	$3s^2S_{1/2}$	0.3	
Rb	37	$5s^{2}S_{1/2}$	30	
Cs	55	$6s^{2}S_{1/2}$	115	
Fr	87	$7s^{2}S_{1/2}$	1100	
T1	81	$6p^{2}P_{1/2}$	-585	
		- /		

An extensive discussion of the significance of the experiment in Ref. [9] for the CP- and T-symmetry violation problem is given in Ref. [1].

We refer the reader to [2] for other proposed experiments on the EDM of paramagnetic atoms: optical pumping of Cs vapor, the use of traps and cooling, and the implantation of Cs into a solid ⁴He matrix. We restrict ourselves to this brief information because our main concern in this paper is with diamagnetic atoms.

1.6 Diamagnetic atoms

The ground state of diamagnetic atoms, i.e., atoms with occupied electron shells, is the state ${}^{1}S_{0}$, and therefore the atomic EDM may be determined only by the nuclear EDM.

Different mechanisms and theoretical models that in principle may be responsible for the existence of EDM in atomic nuclei are discussed in detail in Ref. [1]. Simultaneously discussed is the question of what kind of information and bounds on the constants of the corresponding Hamiltonians may be derived from optical atomic experiments in search of the EDM. Special emphasis is placed on heavy atoms with the nuclear spin I = 1/2, like ¹⁹⁹Hg and ¹²⁹Xe, in particular. Of primary importance in the nuclear EDM of these atoms is the volume effect. The specific cause is the difference between the charge and EDM distributions over the nucleus volume. The magnitude of the corresponding effect is determined by the so-called Schiff moment S and is expressed in units of $e \text{ cm}^3$. For more details about the Schiff moment and its calculation, see Ref. [1]. The expected nuclear EDM of the light ³He atom is far smaller than that of the ¹²⁹Xe and ¹⁹⁹Hg atoms (by many orders of magnitude). As shown by Schiff [7], for the ³He nuclei, the volume effect does not play any part whatsoever. The ³He nuclear EDM is defined by the magnetic interaction of nucleons.

As regards the atomic EDM, the complete screening of the nuclear EDM, which should occur for a point nucleus, is removed if the finite dimension of the nucleus is taken into account. Concerning the relation between the atomic EDM d(A) and the nuclear EDM d(N), see Ref. [1].

The greater the ratio between the nuclear dimensions and the electron shell radii, the greater the nuclear-to-atomic EDM conversion factor i.e., it increases with Z.

Experiments with the ¹⁹⁹Hg atom have advanced the farthest: the first experiment is described in [10], with subsequent improvement in [11]. There are also experiments on the ¹²⁹Xe atomic EDM: the results of the first experiment are outlined in [12]. The Hartree-Fock calculations of the ¹⁹⁹Hg and ¹²⁹Xe atomic EDMs were performed in [13]. An analysis of the first sufficiently reliable experimental data on the ¹⁹⁹Hg and ¹²⁹Xe atomic EDMs showed that optical EDM experiments contain highly valuable information about T-odd effects. The nuclear EDM in suchlike atoms is induced not only by the electric dipole moment of the outer nucleon (neutron) but also, to a large extent, by the PT-odd nucleon-nucleon interaction. As noted in Ref. [5], the nuclear EDM due to this interaction may substantially (up to two orders of magnitude) exceed the EDM of an individual nucleon. This is the reason why the information that may be provided by experiments on the EDM of diamagnetic atoms significantly complements the information acquired from neutron experiments.

The second part of our paper (Sections 2.2, 2.4, 2.5, and 2.6) is dedicated to a possible new experiment on the EDM of atomic ¹²⁹Xe. We therefore limit ourselves to only very brief information about the well-known experiments on the EDM of atomic ¹⁹⁹Hg (the nuclear spin is I = 1/2). In the first experiment by Fortson's group [10], the nuclear spin polarization was produced by optical pumping on the $6^{1}S_{0} \rightarrow 6^{3}P_{1}$ transition involving the intercombination line $\lambda = 254$ nm. The density of spin-polarized atoms was $\sim 10^{13}$ cm⁻³. Measurements were made of the atomic precession frequency shift $\Delta\Omega$ in (1.4) under collinear magnetic and electric fields, i.e., recourse was made to basically the same technique as in the neutron experiment. The precession frequencies were compared in two adjacent cells with a common magnetic field, but with oppositely directed electric fields.

In the second experiment by Fortson's group [11], the $6^1S_0 - 6^3P_1$ transition of atomic ¹⁹⁹Hg was pumped by the

fourth harmonic of a semiconductor laser with the output power 6 mW at $\lambda = 253.7$ nm. Several important technical improvements had been made. As a result, they arrived at the following upper bound for the EDM of atomic ¹⁹⁹Hg (2001):

$$d(^{199}\text{Hg}) < 2.1 \times 10^{-28} e \text{ cm}.$$
 (1.13)

For the calculations of the nuclear EDM of atomic 199 Hg and the nuclear-to-atomic EDM conversion by the Hartree– Fock technique, see Refs [1, 5, 12]. This conversion coefficient does not contain the 'enhancement' factor as for paramagnetic Ti atoms and is determined by the finite nucleus volume, which is small in comparison with the typical atomic dimensions. By ascribing the 199 Hg atomic EDM to the valence neutron in the even–odd nucleus of this atom, it is possible to estimate the upper neutron EDM as

$$d_{\rm n} < 4 \times 10^{-25} \, e \, {\rm cm} \, ,$$

which is worse than result (1.2) of the neutron experiment by nearly an order of magnitude. However, there also exist other interesting possibilities of interpreting the ¹⁹⁹Hg atomic EDM experiment (see Ref. [5]).

The program of experiments to search for and measure the ¹²⁹Xe atomic EDM, which formed beginning with Ref. [13], proceeds from the same basic idea of experiment as the program in Ref. [6] for the neutron EDM and the experiments on the ¹⁹⁹Hg atomic EDM [10, 11]. The spin precession frequency Ω in (1.3) of atomic ¹²⁹Xe in collinear magnetic **B** and electric **E** fields is measured and the shift $\Delta\Omega$ in (1.4), which emerges on reversal of the electric field direction $\mathbf{E} \rightarrow -\mathbf{E}$, is extracted. Naturally, the spin polarization technique is essentially different.

The nuclear spins of rare-gas atoms mixed with rubidium atoms are polarized by way of polarization transfer from optically polarized rubidium atoms in atomic collisions (for more details, see Section 2.1). This is precisely the polarization mechanism that was used in Ref. [13] in the determination of the upper limit for the EDM of ¹²⁹Xe, where the following result (1984) is adduced:

$$d(^{129}$$
Xe) = (0.2 ± 1.1) × 10⁻²⁶ e cm.

The initial version of the experiment has been substantially improved over the past 20 years. The scheme is highly versatile and allows many different realization versions. It permits polarizing the nuclear spins of several rare-gas atoms contained in a common cell under identical conditions, for instance a mixture of ¹²⁹Xe with ¹⁹⁹He. Their anticipated EDM values are radically different. When measuring the quantity $\Delta\Omega$ in (1.4) for atomic ¹²⁹Xe, this allows spin precession frequency measurements for atomic ³He to be used for efficient stability control of the total magnetic field **B**. The stability of the field **B** may be affected by laboratory field fluctuations, as well as by the currents emerging in the reversal of field **E**.

Modern simple and reliable radiation sources for the optical pumping at the resonance transition frequency of rubidium atoms ($\lambda = 795$ nm) — linear semiconductor laser arrays with the output power ~ 15–50 W in the cw mode — enable the operation at high rare-gas densities, up to ten atmospheres [14, 15].

The development of techniques for realizing the maser action mode instead of the free nuclear-spin precession mode is a major accomplishment. With the aid of semiconductor laser pumping, Bear et al. [16] were able to effect the polarization of ³He and ¹²⁹Xe nuclear spins and showed that the ³He and ¹²⁹Xe maser frequency stability was equal to $\sim 10^{-7}$ Hz.

Interestingly, it was not the atomic EDM problem but the CPT-symmetry violation problem that became the motivation for the experiment in Ref. [16]. The latter problem is based on the general principle of local Lorentz invariance, which is common to both the theory of elementary particles and the general theory of relativity. The current status of the Lorentz-invariance violation problem is reviewed in [17]. The possibilities of CPT-symmetry violation are concisely discussed also in Ref. [5].

The main result in Ref. [16] — the high frequency stability of the ³He and ¹²⁹Xe masers at the level of 10^{-7} Hz — is also of major importance for experiments in search of the EDM of atomic ¹²⁹Xe.

Lastly, a simultaneous masing regime of ³He and ¹²⁹Xe was realized with the aid of a two-chamber cell of special geometry, which allowed a high precession frequency stability to be obtained in an experiment to measure the EDM of atomic ¹²⁹Xe [18]. The atoms of ³He and ¹²⁹Xe polarized in the first chamber (the 'pump' chamber) diffuse into the second (maser) chamber. The ³He maser was used as an internal magnetometer, which furnished additional monitoring of the magnetic field and several systematic instrumental effects. This resulted in obtaining the following upper limit for the EDM of ¹²⁹Xe atoms [18], which was five-fold lower than that in Ref. [13]:

$$d(^{129}\text{Xe}) < 4 \times 10^{-27} e \text{ cm}$$
. (1.14)

Possibilities for some other modifications of the measuring technique are discussed in planning new experiments. Exceptionally interesting is the possibility, upon polarizing ¹²⁹Xe atoms, to rapidly cool the sample by optical techniques and transform xenon to the liquid phase with the density $\sim 10^{22}$ cm⁻³. In this case, the polarization persists for a rather long time ($\sim 10^3$ s) and the highest permissible breakdownlimited electric intensity increases to ~ 400 kV cm⁻¹ [19]. According to the author's estimates, the anticipated experimental result for the upper bound of ¹²⁹Xe EDM will be equal to $\sim 10^{-31} e$ cm. It is pertinent to note that Romalis's proposal [19] only appears on the author's site and has nowhere been discussed in detail.

Different methods of recording the precession frequency Ω of atomic nuclear spins are considered in the literature, including the optical recording technique involving the retroaction of spin-polarized rare-gas atoms on Rb. In this case, it is possible to work with both the residual Rb vapor inside the cell (the rubidium density is temperature-controlled) and an Rb magnetometer outside the working volume. A version of this technique was employed in [18]. Despite the wide diversity of well-known projected experiments on the atomic ¹²⁹Xe EDM, their key points are the attainment of the highest possible measurement accuracy for the precession frequency Ω in (1.3) and its shift $\Delta\Omega$ in (1.4) under careful control of the numerous factors that affect the stability of Ω and above all the stability of the magnetic field **B**.

After a major step — moving from a free nuclear spin precession to the maser precession regime (see Refs [16, 18]) — further progress in ¹²⁹Xe EDM experiments in the framework of basically the same scheme is anticipated primarily in technical improvements and careful elaboration of all components and elements of the experiment. It is yet unclear how far it will be possible to advance in this work and whether the purpose at hand is attainable along this path — staging an EDM experiment with an accuracy of about $2 \times 10^{-28} e$ cm, i.e., an accuracy an order of magnitude higher than that in Ref. [18].

In this connection, it is pertinent to discuss the possibility of abandoning the basic scheme of the previous and projected atomic ¹²⁹Xe EDM experiments: moving to recording the electric field of the EDM of spin-polarized ¹²⁹Xe atoms instead of measuring the precession frequency Ω in (1.3) with high precision and extracting the shift $\Delta\Omega$ in (1.4).

A discussion of the possible realization of this approach constitutes the bulk of our paper. We consider a method that involves recording the quadratic Stark effect on the Rydberg levels of an alkali atom, which is proportional to the square of the total electric field $(\mathbf{E}_0 + \mathbf{E}_{\text{EDM}})^2 \approx \mathbf{E}_0^2 + 2\mathbf{E}_0\mathbf{E}_{\text{EDM}}$, where \mathbf{E}_0 is the external field and \mathbf{E}_{EDM} is the field of spin-polarized ¹²⁹Xe atoms. The Stark effect on Rydberg levels exhibits an enhanced sensitivity to the electric field. This gives rise to the effect of specific 'multiplication' of the quantity being measured.

An important feature of the proposed technique is the fact that it does not require high-precision measurements of the nuclear spin precession frequency Ω . As a consequence, the extremely stringent requirements on the stability of the magnetic field **B** are relaxed (see Ref. [6]). The focus of the experiment shifts to the accuracy of frequency measurements of the Stark shift of the Rydberg levels of rare-gas atoms in the optical frequency range.

The proposed method will encounter difficulties of its own, but they are essentially different from those of Ref. [6] and Refs [13, 18]. The method is outlined below (see Sections 2.2, 2.4, 2.5, and 2.6).

2. Search for the electric dipole moment of atomic ¹²⁹Xe

In this section, we propose and discuss a new possible method of searching for the EDM of atomic ¹²⁹Xe. It appears reasonable to provide a brief introductory Section 2.1 dedicated to the method of optical spin polarization of raregas atoms.

2.1 Optical technique for the spin polarization of rare-gas atoms

Production of the polarization of rare-gas atoms (polarization of the nuclear spin I) is of interest in precision experiments aimed at solving several basic problems in modern physics. The development of techniques for obtaining spin-polarized nuclei has a long history.

Perhaps one of the first problems that compelled researchers to pay close attention to the optical method of gaseous target polarization was that of producing an efficient polarizer of thermal neutrons — a target of spin-polarized ³He [20, 21]. The neutrons are polarized because the polarized nuclei of ³He exhibit resonance absorption of neutrons whose spins are antiparallel to the ³He nuclear spins. For a neutron energy of ~ 0.02 eV, the cross section of this resonance absorption is $\sigma_{res} \sim 5.4 \times 10^{-21} \text{ cm}^2$. With an increase in the neutron energy, the cross section σ_{res} decreases in inverse proportion to the neutron velocity. However, for energies

below 10 eV, it remains several thousand times larger than the absorption cross section for neutrons whose spins are parallel to the 3 He nuclear spins.

A universal optical method that enables polarizing any rare-gas atoms with a nonzero nuclear spin (³He, ²¹Ne, ⁸³Kr, ¹²⁹Xe, ¹³¹Xe) had been tested long before the development of present-day laser technology. Of interest in the EDM problem are only the ³He and ¹²⁹Xe atoms, whose nuclear spin is I = 1/2. Added to a rare gas B is an alkali metal A in small amounts of ~ 10¹³-10¹⁴ cm⁻³. The electron spin polarization of A atoms is produced due to the optical pumping by circularly polarized resonance radiation (the ²S_{1/2} \rightarrow ²P_{1/2} transition, D₁ line)

$$\mathbf{A} + n\hbar\omega \to \mathbf{A}\uparrow \quad . \tag{2.1}$$

Subsequently, in collisions of A atoms with B atoms, the electron spin polarization is transferred to the spin of nucleus B:

$$A\uparrow +B \to A + B\uparrow$$
 . (2.2)

This method came into wide use after the advent of lasers with a sufficiently high output power, making it possible to realize efficient optical pumping of rubidium and potassium atoms, Rb atoms first and foremost. A significant part in the development of this method was played by the works of Happer's group [20, 21] (polarization of ³He) and Fortson's group [13] (polarization of ¹²⁹Xe) (for papers that date back to the pre-laser period and the history of research into spin-polarized nuclei production, see Refs [22–24]).

In the first works, the optical pumping of Rb atoms was effected with a dye laser pumped by krypton or argon ion lasers, which provided the pump power ~ 1 W within the D₁ line profile of Rb. Changing to titanium–sapphire lasers enabled increasing the pump power to $\sim 3-4$ W. A further increase in the optical pump power became possible due to the use of linear semiconductor laser arrays in which every laser diode is tuned to the D₁ line of Rb. Using such a linear semiconductor array allowed raising the pump power to 10-50 W. Therefore, work with relatively large volumes and densities of rare-gas atoms became possible.

The spectral width of the emission line of a linear semiconductor array significantly (approximately by an order of magnitude) exceeds the spectral width of the Rb absorption line broadened by an extraneous gas (with the total pressure up to ~ 10 atm). This calls for certain changes in the experimental technique (see, e.g., Refs [14, 15]). Pumping has been done for a large optical thickness of rubidium vapor in the vicinity of the D₁ line. This permits utilizing a substantial part of the absorption profile wings for pumping and, as a consequence, about 80% of the output power of the semiconductor array for the spectral width of its laser line of 2.4 nm.

A large spectral width of the D_1 line broadened by an extraneous gas permits totally neglecting the effects such as the hyperfine structure and the isotopic composition (in the case of Rb, the natural mixture of ⁸⁵Rb and ⁸⁷Rb). This is accounted for in Fig. 1, which serves to conceptually illustrate the spin polarization of atoms by the optical pumping of Rb.

The magnetic sublevels with m = +1/2 and m = -1/2 in Fig. 1 correspond to the quantization axis aligned with the wave vector **k** of the laser beam. The laser radiation with $\lambda = 795$ nm is circularly polarized such that absorption is



Figure 1. Schematic diagram showing the spin polarization of atomic rubidium by the optical pumping technique.

effected only in the state with m = -1/2. The collisions of Rb atoms with each other and with the extraneous gas efficiently 'mix' the states m = -1/2 and m = +1/2. Both the collisions and radiative decay of the excited state ${}^{2}P_{1/2}$ therefore result in the population of the states with m = -1/2 and m = 1/2 of the ground level ${}^{2}S_{1/2}$ with equal probability, while the absorption of circularly polarized radiation depopulates only the m = -1/2 sublevel, as shown in Fig. 1. The constant pumping has the effect that Rb atoms accumulate in the state ${}^{2}S_{1/2}$, m = +1/2, i.e., the electron Rb spin is polarized along the wave vector k. Papchenko, Sobel'man, and Yukov [25] have discussed different mechanisms of Rb spin depolarization in atomic collisions (see also Refs [14, 15]). Under the experimental conditions in Refs [20, 21], collisions between two polarized Rb atoms turn out to be a possible process, and one of the most important ones, leading to Rb spin depolarization:

$$\mathbf{Rb}\uparrow + \mathbf{Rb}\uparrow \to \mathbf{Rb}\uparrow + \mathbf{Rb}\downarrow, \qquad (2.3)$$

with the magnetic dipole interaction being responsible for this process. Chupp et al. [20] give the rate coefficient for this Rb spin relaxation,

$$\langle v\sigma \rangle = 0.8 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1},$$
 (2.4)

for the rubidium density 10^{15} cm⁻³, which is the highest density for this kind of experiment, while the relaxation rate is $N_{\rm Rb} \langle v\sigma \rangle \sim 10^3 \text{ s}^{-1}$.

The formation of Van der Waals Rb₂ molecules in ternary collisions Rb↑+Rb↑+He can also make a contribution to the Rb spin depolarization, depending on the pressure and the external magnetic field. The state Rb₂(${}^{3}\Sigma_{u}$) may also be approximately classified with the type-c Hund coupling, when the spin is 'not attached' to the axis of the molecule and its rotational levels are split by a weak spin-axis interaction. The Rb₂(${}^{3}\Sigma_{u}$) molecular lifetime in a dense gas is short, with the result that depolarization occurs in accordance with the diffusion law. The magnetic field dependence manifests itself because the magnetic field decouples the spin from the angular momentum (see Ref. [25]).

The photons arising from the ${}^2P_{1/2} \rightarrow {}^2S_{1/2}$ radiative transition are polarized differently than the pump radiation, which can in turn give rise to an additional Rb depolarization mechanism. To suppress this process, nitrogen (N₂) at a pressure of about 100 Torr is added to the working cell. For the Rb ${}^2P_{1/2} \rightarrow {}^2S_{1/2}$ radiative transition, the quenching cross section due to Rb-N₂ collisions is equal to ~ 6 × 10⁻¹⁵ cm²

and makes a contribution of ~ 75 MHz to the linewidth, which exceeds the radiative width by more than a factor of 10. In atomic ¹²⁹Xe EDM experiments [13, 18], the addition of N₂ permits increasing the electric field by breakdown prevention, i.e., it entails an additional positive effect.

The efficiency of polarization transfer of the electron spin **s** of an atom to the nuclear spin **I** of a rare-gas atom in binary collisions is significantly different for ³He and ¹²⁹Xe. The leading part in this process is played by the contact interaction

$$H' \propto \left|\Psi(0)\right|^2 \mathbf{sI}\,,\tag{2.5}$$

where $\Psi(0) = \Psi(R = 0)$ is the wave function of the Rb atomic electron that describes its motion in the field of the nucleus of a rare-gas atom in Rb⁻³He and Rb⁻¹²⁹Xe collisions and is taken for R = 0.

The experimental values of the rate coefficient $\langle v\sigma \rangle$ for the reaction of polarization exchange between the spins **s** and **I** in Rb⁻³He and Rb⁻¹²⁹Xe collisions given in Refs [20, 26], respectively, agree nicely with theoretical estimates made by a semiclassical method [15]: the process

 $Rb\uparrow + {}^{3}He \rightarrow Rb + {}^{3}He\uparrow$

is characterized by the rate coefficient

$$\langle v\sigma \rangle = 1 \times 10^{-19} \text{ cm}^3 \text{ s}^{-1}, \qquad (2.6)$$

the process

 $Rb\uparrow + {}^{129}Xe \rightarrow Rb + {}^{129}Xe\uparrow$

is characterized by the rate coefficient

$$\langle v\sigma \rangle = (1-2) \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}.$$
 (2.7)

Therefore, due to interaction (2.5), the nuclear spin is polarized 10^3 times more efficiently in collisions of polarized Rb atoms with atomic ¹²⁹Xe than in Rb⁻³He collisions. This is attributable to the higher charge of the ¹²⁹Xe atomic nucleus (Z = 54), which 'attracts' the wave function of the valence electron of Rb.

In addition to the binary $Rb^{-129}Xe$ collisions, the polarization transfer from the rubidium electron spin s to the nuclear spin I of atomic ¹²⁹Xe can occur during the production of a Van der Waals molecule in $Rb^{-129}Xe$ collisions in the presence of a third body [24]. The ratio between this reaction channel and the binary collision channel depends on the density of ¹²⁹Xe atoms and N₂ molecules, which play the part of the third body. When the 'third body' density is sufficiently high, the efficiency of polarization transfer to the nuclear spin in the case of ¹²⁹Xe can be 10⁴ times higher than in the case of ³He. The atoms of rubidium and helium cannot form a Van der Waals molecule.

The nuclear spin-polarization relaxation processes of ³He and ¹²⁹Xe also differ greatly and depend heavily on specific experimental conditions. Of vital importance in ³He and ¹²⁹Xe nuclear spin relaxation are factors like the purification efficiency of the working substances, the material and kind of processing of the walls of the working cell, the specially applied coatings, etc. We therefore give only some of the averaged characteristics and parameters of experiments; for more detailed information, we refer the reader to the papers cited above. Upon turning off the pump, the nuclear polarization of ³He persists for a rather long time (from several hours to dozens of hours). For ¹²⁹Xe, this period is

much shorter $(10^3 - 10^4 \text{ s})$.

The composition of the working mixture is varied, depending on the goals of the experiment. In particular, in an experiment in search of the EDM of atomic ¹²⁹Xe [18], use was made of the following working mixture: rubidium vapor with the density 2×10^{13} cm⁻³, xenon (90 % enriched with the ¹²⁹Xe isotope), ³He, and N₂ at the respective partial pressures 110, 1115, and 80 Torr.

Employing a semiconductor laser array with the output power $\sim 15-50$ W as the source of optical pumping enables the realization of an approximately 50 % polarization of ³He atoms with the density $\sim 10^{20}$ cm⁻³ and of ¹²⁹Xe atoms with the density $\sim 10^{19}$ cm⁻³.

In concluding this section, we note that recent years have unexpectedly seen the emergence of a new promising area involving the use of rare gases with polarized nuclei in medicine. It has been shown that inhaling polarized ³He rare gas and especially ¹²⁹Xe enables the diagnosis of lungs by NMR tomography (see, e.g., Refs [27, 28]).

2.2 Electric field of the EDM of spin-polarized ¹²⁹Xe atoms

The electric field produced by the polarization **P** of a unit volume in an unbounded medium can be calculated by a method elaborated in the microscopic dispersion theory [29]. In the case of a finite volume, calculating this field inside and outside the volume requires a special consideration for every specific volume shape — sphere, cylinder, etc. For definiteness, we consider a dielectric sphere. It may be uniformly polarized by an external field **G**, as well as by coercive orientation of constant (independent of **G**) dipoles inside the sphere. The polarization **P** produced by the EDM of spinpolarized atoms ¹²⁹Xe can be represented as polarization due to external currents in the framework of macroscopic electrodynamics, just as the polarization is treated in the case of piroelectrics.

We consider a dielectric sphere in a uniform electric field **G**, following Landau and Lifshits [30, p. 63, 64] and Tamm [31, § 24]. We define the field potentials φ and intensities $\mathbf{E} = -\nabla \varphi$ inside (*i*) and outside (*e*) the sphere as

$$\varphi^{(i)} = -\mathbf{G}\mathbf{R} + \mathbf{F}\mathbf{R}, \quad \mathbf{E}^{(i)} = \mathbf{G} - \mathbf{F}, \qquad (2.8)$$

$$\varphi^{(e)} = -\mathbf{G}\mathbf{R} + \left(\frac{R_0}{R}\right)^3 \mathbf{F}\mathbf{R},$$

$$\mathbf{E}^{(e)} = \mathbf{G} - \left(\frac{R_0}{R}\right)^3 \left[\mathbf{F} - 3\mathbf{n}(\mathbf{n}\mathbf{F})\right],$$
(2.9)

where R_0 is the sphere radius, $\mathbf{n} = \mathbf{R}/R$, and \mathbf{F} is an arbitrary vector, which is noncollinear with \mathbf{G} in general and is determined by the boundary conditions at the sphere surface.

The boundary conditions at the sphere surface $\mathbf{R} = \mathbf{R}_0$ for the potentials $\varphi^{(i)}(\mathbf{R}_0) = \varphi^{(e)}(\mathbf{R}_0)$ are automatically satisfied by relations (2.8) and (2.9). It is also necessary to satisfy the continuity condition for the normal projection of the induction vector **D**,

$$\mathbf{R}_0 \mathbf{D}^{(i)} = \mathbf{R}_0 \mathbf{D}^{(e)} \,.$$

Because

$$\mathbf{R}_{0}\mathbf{D}^{(e)} = \varepsilon^{(e)}\mathbf{R}_{0}\mathbf{E}^{(e)} = \varepsilon^{(e)}\mathbf{R}_{0}\mathbf{G} - \varepsilon^{(e)}\mathbf{R}_{0}\left[\mathbf{F} - 3\mathbf{n}(\mathbf{n}\mathbf{F})\right]$$
$$= \varepsilon^{(e)}\mathbf{R}_{0}\mathbf{G} + 2\varepsilon^{(e)}\mathbf{R}_{0}\mathbf{F},$$

$$\mathbf{R}_0 \mathbf{D}^{(i)} = \varepsilon^{(e)} \mathbf{R}_0 (\mathbf{3G} - \mathbf{2E}^{(i)})$$

or

$$\mathbf{R}_0 \left(\mathbf{D}^{(i)} + 2\varepsilon^{(e)} \mathbf{E}^{(i)} - 3\varepsilon^{(e)} \mathbf{G} \right) = 0.$$

In view of the arbitrariness of \mathbf{R}_0 , it is possible to derive the following important relation between the vectors $\mathbf{E}^{(i)}$ and $\mathbf{D}^{(i)}$ inside the sphere [see formula (8.1) in Ref. [30]]:

$$\mathbf{D}^{(i)} + 2\varepsilon^{(e)}\mathbf{E}^{(i)} = 3\varepsilon^{(e)}\mathbf{G}.$$
(2.10)

In formulas (2.8)–(2.10), $\varepsilon^{(i)}$ and $\varepsilon^{(e)}$ are the respective permittivities inside and outside the sphere.

Formula (2.10) is universal. No assumptions about the relation between $\mathbf{D}^{(i)}$ and $\mathbf{E}^{(i)}$ have been made. In particular, no assumptions about a linear relation between $\mathbf{D}^{(i)}$ and $\mathbf{E}^{(i)}$ and about their collinearity have been made. This last statement is associated with the consideration in Ref. [31] extended in comparison with Ref. [30].

As in the case of piroelectrics (see Ref. [30, p. 85-87]), we assume that

$$\mathbf{D}^{(i)} = \varepsilon^{(i)} \mathbf{E}^{(i)} + 4\pi \mathbf{P}.$$

Without the term $4\pi \mathbf{P}$, this is the standard relation between $\mathbf{D}^{(i)}$ and $\mathbf{E}^{(i)}$ in an isotropic dielectric. The term $4\pi \mathbf{P}$ gives the polarization produced by external currents, which are in this case the EDM of spin-polarized ¹²⁹Xe atoms,

$$\mathbf{P} = N\mathbf{d}\,,\tag{2.11}$$

where N is the density of spin-polarized ¹²⁹Xe atoms and **d** is the ¹²⁹Xe atomic EDM.

It follows from (2.10) and (2.11) that

$$(\varepsilon^{(i)} + 2\varepsilon^{(e)}) \mathbf{E}^{(i)} + 4\pi \mathbf{P} = 3\varepsilon^{(e)}\mathbf{G}$$

or

$$\mathbf{E}^{(i)} = \frac{3\varepsilon^{(e)}\mathbf{G}}{\varepsilon^{(i)} + 2\varepsilon^{(e)}} - \frac{4\pi\mathbf{P}}{\varepsilon^{(i)} + 2\varepsilon^{(e)}} \,. \tag{2.12}$$

For $\varepsilon^{(e)} = \varepsilon^{(i)} = 1$, we have

$$\mathbf{E}^{(i)} = \mathbf{G} - \frac{4\pi}{3} \mathbf{P}.$$

In expression (2.9) for the field $\mathbf{E}^{(e)}$ outside the sphere, we replace the vector \mathbf{F} by $\mathbf{G} - \mathbf{E}^{(i)}$ to obtain

$$\mathbf{E}^{(e)} = \mathbf{G} - \frac{\varepsilon^{(i)} - \varepsilon^{(e)}}{\varepsilon^{(i)} + 2\varepsilon^{(e)}} \left(\frac{R_0}{R}\right)^3 \left[\mathbf{G} - 3\mathbf{n}(\mathbf{nG})\right] - \frac{4\pi}{\varepsilon^{(i)} + 2\varepsilon^{(e)}} \left(\frac{R_0}{R}\right)^3 \left[\mathbf{P} - 3\mathbf{n}(\mathbf{nP})\right].$$
(2.14)

The first two terms in (2.14) give the electric field in the case where the sphere is polarized by the external field **G**. The field produced by the polarization **P** outside the sphere in addition to the external field **G** is

$$\mathbf{E}_{\text{EDM}}^{(e)} = -\frac{4\pi}{\varepsilon^{(i)} + 2\varepsilon^{(e)}} \left(\frac{R_0}{R}\right)^3 \left[\mathbf{P} - 3\mathbf{n}(\mathbf{nP})\right].$$
(2.15)

If the vector **R** is orthogonal to **P**, then

$$\mathbf{E}_{\text{EDM}}^{(e)} = -\frac{4\pi \mathbf{P}}{\varepsilon^{(i)} + 2\varepsilon^{(e)}} \left(\frac{R_0}{R}\right)^3.$$
 (2.16)

Therefore, the total field \mathbf{E}_{tot} outside the sphere is (for $\varepsilon^{(i)} = \varepsilon^{(e)} = 1$ and $\mathbf{R} \perp \mathbf{P}$)

$$\mathbf{E}_{\text{tot}} = \mathbf{G} - \frac{4}{3} \pi \left(\frac{R_0}{R}\right)^3 \mathbf{P}.$$
 (2.17)

At a point located at a distance L from the sphere surface in the direction **R** that is short in comparison with R_0 (for instance, $L \le 0.25R_0$), the factor $(R_0/R)^3$ is close to unity:

$$\left(\frac{R_0}{R_0+L}\right)^3 \approx 1-0.5\,.$$

Hence, for such values of L,

$$\mathbf{E}_{\text{tot}} \simeq \mathbf{G} - \frac{4}{3} \,\pi \mathbf{P} \,. \tag{2.18}$$

From expression (2.17), it follows that at distances $R \gg R_0$,

$$\mathbf{E}_{\text{tot}} = \mathbf{G} - \frac{\mathbf{D}_{\text{tot}}}{R^3} \,, \tag{2.19}$$

where \mathbf{D}_{tot} is the total EDM of the sphere equal to $(4/3)\pi R_0^3 N \mathbf{d}$, as expected.

If a magnetic field **B** is imposed perpendicular to the plane (**G**, **P**), owing to the precession of polarized ¹²⁹Xe atomic spins with a frequency Ω , the scalar product **PG** and the projection of the vector **P** on the direction **G** oscillate:

$$P_G = P\cos(\Omega t + \varphi_0), \quad \mathbf{PG} = PG\cos(\Omega t + \varphi_0).$$
 (2.20)

2.3 ns Rydberg states of alkali atoms. The Stark effect

The nl ($l \ge 1$) Rydberg states of alkali atoms are hydrogenlike. The energy of these states is written as

$$E_{nl} \approx -\frac{\mathrm{Ry}}{n^2} \left(1 + 2\frac{\Delta_l}{n}\right),$$
 (2.21)

where $Ry = (1/2)me^4/\hbar^2$ is the Rydberg constant (Ry = 13.5 eV or $2.15 \times 10^{-11} \text{ erg}$) and Δ_l is the quantum defect. The energy splitting of the states *ns*, *np* is

$$\Delta E_{\rm sp} \approx -2 \, \frac{\rm Ry}{n^3} \left(\Delta_{\rm s} - \Delta_{\rm p} \right). \tag{2.22}$$

For all alkali atoms (from Li to Cs), $\Delta_s - \Delta_p = 0.346 - 0.485$. For Rb and Cs atoms, which are mainly considered below,

the respective difference $\Delta_s - \Delta_p$ is 0.485 and 0.477, and

$$\Delta E_{\rm sp} \approx -0.95 \, \frac{\rm Ry}{n^3} \,. \tag{2.23}$$

We also give the value of the matrix element for the electron radius vector r_{sp} and the radiative lifetime *T* of the level with the principal quantum number *n* averaged over all the *l*, *m* sublevels:

$$r_{\rm sp} = \frac{3}{2} n^2 a_0 \,, \tag{2.24}$$

$$T = \frac{1}{3} \left[\frac{n^5}{\ln(n/1,1)} \right] A_0^{-1}, \qquad (2.25)$$

where $A_0 = 7.9 \times 10^9 \text{ s}^{-1}$.

The radiative lifetime of the ns state is different from the averaged one in (2.25). This issue is discussed in Section 2.5.

The ns level shift in the electric field E (quadratic Stark effect) is

$$\Delta E_{ns} = \frac{e^2 r_{sp}^2}{\Delta E_{sp}} E^2 = \frac{9}{4} n^4 \frac{e^2 a_0^2}{\Delta E_{sp}} E^2.$$
(2.26)

This formula applies when $\Delta E_{ns} < \Delta E_{sp}$:

$$\frac{9}{4}n^4e^2a_0^2E^2 < 0.95\,\frac{\mathrm{Ry}^2}{n^6}\,,\tag{2.27}$$

$$n^{10}E^2 < 0.3 \times 10^{14} \,. \tag{2.28}$$

Here, the electric intensity *E* is expressed in CGSE units (1 CGSE unit corresponds to 300 V cm^{-1}) and a_0 is the atomic mass unit.

We consider the Stark effect in the *n*s state of an alkali atom that simultaneously experiences the action of the EDM field of spin-polarized ¹²⁹Xe atoms and the external electric field \mathbf{E}_0 . For definiteness, we assume that the spin-polarized atomic ¹²⁹Xe fills a spherical volume of radius R_0 , the alkali atom is outside this volume, and the external field \mathbf{E}_0 is uniform. The selected configuration allows using formula (2.18) (for $L \leq 0.25R_0$):

$$\mathbf{E}_{\text{tot}} = \mathbf{E}_0 - \frac{4\pi}{3} \,\mathbf{P} \,. \tag{2.29}$$

We set

$$\mathbf{P} = N\mathbf{d}\,,\tag{2.30}$$

where N is the number of spin-polarized 129 Xe atoms in 1 cm³ and **d** is the sought EDM. It is taken as

$$d = 2 \times 10^{-28} e \text{ cm} \approx 10^{-37} \text{ CGSE},$$
 (2.31)

which corresponds to the goals of the program to search for the EDM of a neutron [6] and a ¹²⁹Xe atom. Because $E_0 \ge 4\pi Nd$, it follows from formulas (2.26), (2.23), (2.29), and (2.30) that

$$\frac{1}{2\pi} \frac{\Delta E_{ns}}{\hbar} \approx I + \Delta I = -\left(\frac{0.7}{2\pi}\right) \times 10^3 n^7 \left\{ E_0^2 - \frac{8\pi}{3} N \mathbf{E}_0 \mathbf{d} \right\}.$$
(2.32)

The quantity $\Delta I \propto d$ dependent on the sought EDM is

$$\Delta I = \frac{5.8}{2\pi} \times 10^3 n^7 N \mathbf{E}_0 \mathbf{d} \,. \tag{2.33}$$

For convenience of comparing ΔI with the quantity $4\mathbf{E}_0 \mathbf{d}/\hbar$, which is the main characteristic of the sought EDM effect in formula (1.4), we rewrite expression (2.33) as

$$\Delta I = \frac{1.5}{2\pi} \times 10^{-4} n^7 \left(\frac{N}{10^{20}}\right) \times 4 \frac{\mathbf{E}_0 \mathbf{d}}{\hbar} .$$
 (2.34)

Here, we used that large values of N of the order $10^{19} - 10^{20}$ cm⁻³ are considered below; ΔI is expressed in Hertz.

For $N = 10^{20}$ cm⁻³ and the value of *d* according to assumption (2.31), the ratio between ΔI and the main term *I*

in expression (2.32) is

$$\frac{\Delta I}{I} = \frac{8\pi}{3} \; \frac{NE_0 d}{E_0^2} \approx 0.8 \times 10^{-16} \; \frac{1}{E_0} \; . \tag{2.35}$$

From inequality (2.28), it follows that the quadratic Stark effect of the level *ns* is described by formulas (2.26), (2.32), (2.34), and (2.35) provided that inequality (2.28) is satisfied, which is the case for the mutually related *n* and E_0 values collected in Table 2 below. Also given in Table 2 are the values of ΔI , Eqn (2.34), and the ratio $\Delta I/I$ in (2.35) for each possible pair of the parameters *n* and E_0 . It is assumed that $N = 3 \times 10^{19}$ cm⁻³. In accordance with (2.28), the greatest admissible values are $E_0 \propto 1/n^5$ and $\Delta I \propto n^2$. It is noteworthy that going beyond the limits of the quadratic Stark effect is undesirable because it is of significance in the subsequent discussion that the quantity under measurement is proportional to $\mathbf{E}_0 \mathbf{E}_{\text{EDM}}$ and not merely to \mathbf{E}_{EDM} , as it is under the quadratic Stark effect.

Referring to Table 2, the values in the last three columns (the values $n \sim 100-200$) show the greatest promise for the proposed experiment. The lowering of the field E_0 required to satisfy Eqn (2.21) is overcompensated for by the factor n^7 in formula (2.34). The quantity ΔI under measurement, from which it is possible to determine the EDM *d* for n = 100 and n = 200, is approximately 2×10^4 times greater than the quantity $\Delta \Omega/2\pi = 2 \times 10^{-8}$ Hz in (1.10). In this case, the ratio $\Delta I/I$ is comparable in magnitude with $\Delta \Omega/\Omega \approx 2 \times 10^{-11}$ Eqn (1.9). The next section is concerned with the basic scheme of the experiment in which it is possible to measure the Stark effect of Rydberg atoms in electric field (2.22).

Table 2.

n	20	100	150	200
<i>E</i> ₀ , CGSE, below	1.7	5.5×10^{-4}	6.6×10^{-5}	1.7×10^{-5}
E_0 , V cm ⁻¹ , below	500	1.7×10^{-1}	2×10^{-2}	$5 imes 10^{-3}$
$\Delta I/I$	1.6×10^{-17}	6.7×10^{-14}	$3.3 imes 10^{-13}$	$1.7 imes 10^{-12}$
ΔI , Hz	6.7×10^{-6}	$1.6 imes 10^{-4}$	3.3×10^{-4}	$6.7 imes 10^{-4}$

2.4 Basic scheme of the experiment on the Stark shift of *n*s Rydberg levels of alkali atoms

The following basic experiment scheme is appropriate for observing the Stark effect with an alkali atom in the *n*s Rydberg state, which simultaneously experiences the EDM field of spin-polarized ¹²⁹Xe atoms and the external electric field \mathbf{E}_0 (Fig. 2). A collimated beam of alkali atoms traveling with a velocity **v** is formed near the surface of a volume containing spin-polarized ¹²⁹Xe and ³He atoms, and the electric field \mathbf{E}_0 is imposed parallel to **v**. In the region of the closest approach of the beam to the volume of spin-polarized ¹²⁹Xe and ³He atoms, the atoms of the beam are laser-excited to one of the states *n*s with $n \sim 100-200$ (the exact value of *n* is insignificant). Upon transiting the excitation region, the beam of alkali atoms is recorded with an ionization detector, which is responsive only to highly excited Rydberg atoms.

In a cell with the polarized mixture of 129 Xe and 3 He, the precession of nuclear spins is initiated in the magnetic field **B**



Figure 2. Basic schematic of the experiment.

applied perpendicular to the plane $(\mathbf{E}_0, \mathbf{P})$. It is assumed that the ¹²⁹Xe atom has an EDM and the ³He atom does not. The spin precession of ¹²⁹Xe should give rise to a variable electric field \mathbf{E}_{EDM} alternating at a frequency Ω , which is planned to be measured from the Stark shift of the ns Rydberg level of the alkali atom. The flow of excited atoms some distance away from the cell is recorded by the ionization detector, whose signal is used to automatically lock the laser frequency to the slope of the excitation line profile of the *n*s level via a relatively slow feedback loop. The variation in the transition wavelength arising from the variation of the applied electric field $\mathbf{E}_0 + \mathbf{E}_{\text{EDM}}$ is the measure of the *n*s level Stark shift. The contribution of the term $\Delta I \propto E_0 E_{EDM}$ in (2.34) is extracted from the detector signal at a relatively high frequency Ω of the ¹²⁹Xe atomic spin precession. The signal at the ³He precession frequency is then used to monitor spurious effects. The measurement procedure is calibrated by changing the electric field E_0 by a known value.

Rb and Cs alkali atoms are suitable candidates for the role of a Rydberg atom. Two-photon excitation involving counterpropagating laser beams, which minimizes the contribution of the Doppler effect, appears to be most attractive for the excitation of the *ns* levels. The two-photon excitation $n_0 s \rightarrow ns$ (n_0 is the ground state, $n \sim 100-200$) necessitates lasers with the respective laser wavelengths $\lambda \sim 0.59 \ \mu\text{m}$ and $\lambda \sim 0.63 \ \mu\text{m}$. But this turns out to be hardly possible. In both cases, the nearest virtual level $\tilde{n}p$ is located such that the resonance defect $E_{n_0 s} + \hbar \omega - E_{\bar{n}p}$ is too large, of the order of $(0.1-0.05) \ \text{Ry} \sim 10^{-12} \ \text{erg}$. As a consequence, the twophoton excitation cross section, which is approximately equal for Rb and Cs, is small:

$$\sigma_{n_0 s \to n s} \sim 10^{-39} \text{ cm}^4 \text{ s}$$
. (2.36)

For a cw single-frequency laser, it is expedient to rely on an intensity F of the order of $10^{17} - 10^{18}$ photon cm⁻² s⁻¹. The excitation probability is then given by $W = \sigma F^2 \sim 10^{-3} \text{ s}^{-1}$ (we recall that an atom resides in the laser beam during its transit time of the order 10^{-4} s). According to the above estimates, two-photon excitation is unrealistic in the experiment under discussion. For an alternative, there is good reason to consider a twostage scheme, $n_0 s \rightarrow n_1 p \rightarrow ns$. The choice of a specific scheme and suitable lasers for stepwise excitation is governed by the necessity to afford a reasonable signal acquisition time required for averaging statistical errors.

This problem is discussed in Section 2.5. We show there that a realization of the final excitation stage $n_1 p \rightarrow ns$ in the infrared range (10.6 or ~ 100 µm), where the Doppler broadening is much smaller than in the optical range, offers the best advantage. Presently available in the laser oscillation regime are the wavelengths $\lambda = 118.83 \text{ µm}$ (lasing by CH₃OH molecules) and $\lambda = 184.31 \text{ µm}$ (lasing by CH₂F₂ molecules) in the far infrared range for an output power above 100 mW. This is a SIFIR-50 laser system (pumped by a CO₂ laser with wavelength selection) manufactured by Coherent, Inc. The GEM Select 50 laser furnishes a wide set of laser wavelengths in the 9.174–10.835 µm range for an output power of more than 10 mW.

As in many other experiments involving Rydberg atoms, the problem of thermal wall radiation (of the whole cavity of the experimental volume) emerges. For Rydberg states with $n \sim 100$, the thermal radiation in the spectral range ω , $\omega + \Delta \omega$ is described by the Rayleigh–Jeans law for the photon flux

$$\Delta N_{\omega}^{\rm f} = \frac{\omega \Delta \omega kT}{\pi^2 c^2 \hbar} , \quad \omega \ll \frac{kT}{\hbar} , \qquad (2.37)$$

where k is the Boltzmann constant. In the estimates below, we put t = 300 K and $kT/\hbar \approx 4 \times 10^{13}$ s⁻¹. This brings up the question: Can thermal radiation (2.37) give rise to the ionization of the *n*s state, i.e., to a photoeffect, on the path from the excitation region to the detector and, in addition, to the mixing of the states $n, n + \Delta n$?

We estimate the probability $W(s^{-1})$ of both processes. In the Kramers approximation, the effective photoeffect cross section σ_n^{ph} for a level *n* averaged over all *l* sublevels is (see, e.g., Ref. [32]) is given by

$$\sigma_n^{\rm ph} = \frac{64\pi}{3\sqrt{3}} \,\alpha \left(\frac{\rm Ry}{\hbar\omega}\right) \frac{a_0^2}{n^5} \,, \tag{2.38}$$

where $\alpha = e^2/\hbar c = 1/137$. For an estimate, it may be assumed that $\omega = \text{Ry}/n^2\hbar = 2 \times 10^{16}/n^2$ (photoelectric threshold) and $\Delta \omega \simeq \omega/2$. As a result, we obtain

$$W^{\rm ph} = \Delta N_{\omega}^{\rm ph} \sigma_n^{\rm ph} \simeq \frac{0.7 \times 10^6}{n^3} \, [{\rm s}^{-1}] \,.$$
 (2.39)

For $n \sim 100$, the probability $W^{\rm ph} \sim 1 \, {\rm s}^{-1}$. On the path length *L* from the excitation region to the detector, i.e., in a time $\tau = L/v$ known not to exceed 10^{-2} s, we have $W^{\rm ph}\tau < 10^{-2}$. This implies that the photoionization of the *n*s states, $n \sim 100$, due to the thermal wall radiation can be neglected.

We also estimate the $n, n \pm \Delta n$ state mixing due to thermal radiation (2.37). To the $ns \rightarrow np$ transition, there corresponds the effective cross section

$$\sigma_{ns,np}^{ph} = \frac{\lambda^2 A}{4\Delta\omega} , \qquad (2.40)$$

where λ is the wavelength,

$$\frac{2\pi c}{\omega} = \frac{2\pi c\hbar}{\mathrm{Ry}} n^3,$$

A is the $np \rightarrow ns$ spontaneous radiative transition probability, and $\Delta \omega$ is the width of absorption spectrum:

$$A = \frac{4\omega^3}{3\hbar c^3} \left| e \mathbf{r}_{ns,np} \right|^2.$$
(2.41)

For the hydrogen atom, the matrix element $\mathbf{r}_{ns,np}$ can be calculated exactly. The radial integral [see formula (2.24)] is $r_{ns,np} = (3/2)a_0n^2$, and therefore, for hydrogen-like states with $n \sim 100$,

$$A = 24\pi^3 \left(\frac{a_0}{\lambda}\right)^3 \left(\frac{e^3}{a_0\hbar}\right) n^4 \simeq 0.7 \times 10^{10} \frac{1}{n^5} \, [\mathrm{s}^{-1}]. \quad (2.42)$$

As a result, we obtain

$$W(ns, np) = \Delta N_{\omega}^{ph} \sigma_{ns, np}^{ph} \simeq 10^7 \frac{1}{n^2} [s^{-1}].$$
 (2.43)

The probabilities of the transitions $ns \rightarrow (n + \Delta n)$ p, $\Delta n = 1, 2, ...,$ may be estimated by the Kramers formula for the $n + \Delta n \rightarrow n$ spontaneous radiative transition probability averaged over the *l*, *l'* sublevels of the levels *n* and $n + \Delta n$ (see formula (9.337) in Ref. [32]):

$$A_{n+\Delta n,n} = 1.2 \times 10^{10} \, \frac{1}{n^5 \Delta n} \, [\mathrm{s}^{-1}] \,. \tag{2.44}$$

For an estimate, we put $A_{(n+\Delta n)p,ns} \simeq A_{n+\Delta n,n}$ from formula (2.44).

For $\Delta n = 1$, quantities (2.42) and (2.44) are about the same in magnitude, which permits using approximation (2.44). On multiplying $\Delta N_{\omega}^{\rm ph}$ from formula (2.37) by the cross section

$$\sigma_{ns,(n+\Delta n)p}^{\text{ph}} = \frac{\lambda^2 A_{n+\Delta n,n}}{4\Delta\omega}$$

the spectral widths are canceled. For the $n \rightarrow n + \Delta n$ transition probability due to the action of thermal radiation, we eventually obtain

$$W(ns, (n + \Delta n) p) \simeq 1.2 \times 10^7 \frac{1}{n^2 (\Delta n)^2} [s^{-1}].$$
 (2.45)

Formula (2.45) can be applied for both $\Delta n > 0$ and $\Delta n < 0$ ($\Delta n = 1, 2, ...$), which corresponds to stimulated emission instead of absorption.

For $n \sim 100$, probabilities (2.43) and (2.45) are of the respective orders 10^3 s^{-1} and $10^3 \times 1/\Delta n^2 \text{ s}^{-1}$. Because of the factor $1/\Delta n^2$, the *n*s state mixing with the neighbors $n \pm 1$, $n \pm 2, \ldots$ due to the thermal wall radiation on the path from the excitation region to the detector extends to only its closest neighbors and is therefore insignificant in the measurement version being proposed. This is an important special feature of the experiment scheme under discussion, because the flux of excited atoms is recorded without attributing them to specific *n* or *l*.

2.5 Statistical errors

As in the neutron experiment, in the method under discussion, it is necessary to extract the desired effect $\Delta I \propto \mathbf{E}_0 \mathbf{d}/\hbar$ in (2.34) from the background of statistical errors δI_{st} and systematic instrumental errors δI_{instr} . In this section, we consider the statistical noise arising from the Poisson distribution of the flux $N_{ns}(L)$ of excited alkali atoms in the *n*s state that reach the ionization detector (*L* is the detector distance from the *ns*-state excitation region). To average this noise to a level permitting the desired effect ΔI to be recorded, the signal acquisition time *T* defined by the relation

$$\frac{\Delta I}{\Delta \omega / 2\pi} = \frac{1}{\sqrt{N_{ns}(L) T}}$$
(2.46)

is required. Here, $\Delta \omega$ is the width of the *ns*-state excitation spectrum.

In the case of a two-photon excitation $n_0 s \rightarrow ns$ with counterpropagating beams, the magnitude of $\Delta \omega$ is determined by the time-of-flight broadening $\Delta \omega_{\tau} = 1/\tau$. For the Rb and Cs alkali atoms adopted above, using a two-photon excitation encounters difficulties due to the smallness of the effective cross section of this process $\sigma(n_0 s \rightarrow ns) \sim$ 10^{-39} cm⁴ s (see Section 2.4). That is why we consider only the two-stage excitation $n_0 s \rightarrow n_1 p \rightarrow ns$ below. In this case, $\Delta \omega$ is the Doppler width,

$$\frac{\Delta\omega_{\rm D}}{2\pi} = \frac{\omega v_{\perp}}{2\pi c} = \frac{v_{\perp}}{\lambda} \; ,$$

where v_{\perp} is the transverse velocity component of an alkali atom (the projection on the laser beam direction) and λ is the $n_1 p \rightarrow ns$ transition wavelength.

As a result, the left-hand side of (2.46) assumes the form $\Delta I\lambda/v_{\perp}$. Setting $\Delta I = 10^{-3}$ Hz (see Table 2), for $v_{\perp} \sim 10^{-3} v \approx 10$ cm s⁻¹ we obtain

$$T \sim \left(\frac{10^4}{\lambda}\right)^2 \frac{1}{N_{ns}(L)} \, [s^{-1}],$$
 (2.47)

where λ is expressed in centimeters.

If the $n_1 p \rightarrow ns$ transition corresponds to the optical spectral region, $\lambda \sim (0.5-1) \times 10^{-4}$ cm, setting $N_{ns}(L) \sim 10^8 \text{ s}^{-1}$, we obtain

 $T\sim 10^8$ s.

The situation may be substantially improved if infrared lasers are used for the excitation of the final stage transition. In accordance with expression (2.47), for $N_{ns}(L) = 10^8 \text{ s}^{-1}$,

$$T \sim \begin{cases} 10^6 \text{ s for } \lambda \approx 10 \ \mu\text{m} \,, \\ 10^4 \text{ s for } \lambda \approx 100 \ \mu\text{m} \,. \end{cases}$$
(2.48)

For $\lambda \approx 100 \ \mu\text{m}$, the Doppler width of the excitation line reaches the level of the transit linewidth, and a further increase in the $n_1\text{p} \rightarrow n\text{s}$ transition excitation wavelength would therefore not serve the purpose of decreasing $\Delta\omega_{\text{D}}$.

To excite the $n_1 p \rightarrow ns$ transition with a $\lambda \approx 10 \ \mu m$ laser requires effecting the excitation from an $n_1 p$ level, $n_1 \approx 14-17$; to excite the $n_1 p \rightarrow ns$ transition with a $\lambda \approx 100 \ \mu m$ laser requires levels with $n_1 \approx 33-38$. The aforesaid equally applies to Rb and Cs atoms.

For definiteness, we primarily consider the Cs atom in the subsequent discussion. It seems expedient to ensure an efficient $n_1p \rightarrow ns$ excitation of one of the *ns* Rydberg states using a SIFIR-50 laser system (Stabilized Integrated laser system for the Far InfraRed wavelength range) in the far-IR spectral range: at the wavelengths $\lambda = 118.83 \mu m$ (singlefrequency lasing by CH₃OH molecules) and $\lambda = 184.34 \mu m$ (single-frequency lasing by CH₂F₂ molecules). These molecules are pumped by a CO₂ laser with wavelength selection. The SIFIR-50 system provides the output power above 100 mW at both wavelengths, while the spectral drift of the free-running laser line does not exceed 100 kHz over a 15-second period.

The first excitation stage $6s \rightarrow n_1p$, $n_1 = 34$ or 35, $\lambda \sim 0.34 \mu m$, may be effected by doubling the frequency of a dye or titanium-sapphire laser.

The flux $N_{ns}(L)$ is conveniently expressed in terms of the total alkali atomic flux N_0 measured in units of s⁻¹,

$$N_{\rm ns}(L) = \beta_1 \beta_2 \beta_3 N_0 \,, \tag{2.49}$$

where β_1 is the efficiency of the first excitation stage $n_0 s \rightarrow n_1 p$, β_2 is the efficiency of the second stage $n_1 p \rightarrow ns$, and β_3 is the flux attenuation factor on the path λ .

Stringent requirements are imposed on the frequency parameters (spectral width, stability) of the last-stage laser. The first-stage laser should provide a radiation intensity F(cm⁻² s⁻¹) such that the absorption probability $\sigma_{\gamma'\gamma}F(s^{-1})$ exceeds the inverse time of flight through the working region $1/\tau$ (s⁻¹) and the radiative decay probability of the level to be populated A_{γ} (s⁻¹).

Under conditions where the Doppler transition width $\Delta \omega_{\rm D}$ exceeds $1/\tau$ and A_{γ} , the absorption cross section is given by the relation

$$\sigma_{\gamma'\gamma} = \frac{\lambda^2}{4} \frac{A_{\gamma\gamma'}}{\Delta\omega_{\rm D}} \approx \frac{\lambda^3 A_{\gamma\gamma'}}{8\pi v_{\perp}} \sim 4 \times 10^{-3} \lambda^3 A_{\gamma\gamma'} \,. \tag{2.50}$$

The quantities $A_{\gamma\gamma'}$ and the corresponding absorption cross sections $\sigma_{\gamma'\gamma}$ for the 6s \rightarrow 35p and 35p \rightarrow *n*s transitions calculated with the aid of FAC [33] are given below:

$$\begin{split} &A(35p^2 P_{1/2} \to 6s^2 S_{1/2}) = 0.65 \times 10^3 \ s^{-1} \,, \\ &\sigma(6s^2 S_{1/2} \to 35p^2 P_{1/2}) = 1.4 \times 10^{-13} \ cm^2 \,, \end{split}$$

$$A(ns^{2}S_{1/2} \to 35p^{2}P_{1/2}) = 1 s^{-1},$$

$$\sigma(35p^{2}P_{1/2} \to ns^{2}S_{1/2}) = 5 \times 10^{-9} cm^{2}.$$
(2.52)

We assume that the transition via the intermediate state 35p is effected via the ${}^{2}P_{1/2}$ level and not the ${}^{2}P_{3/2}$ level. For $F \sim 10^{18}$ cm⁻² s⁻¹, the conditions

$$F\sigma(6s^2S_{1/2} \to 35p^2P_{1/2}) \gg A(35p^2P_{1/2} \to 6s^2S_{1/2}), \ \frac{1}{\tau}$$

hold, which, in view of the induced $35p \rightarrow 6s$ transitions (the saturation condition) ensure the relation

$$N_{6s}(\text{out}) = N_{35p}(\text{out}) = \frac{1}{2}N_0, \quad \beta_1 = \frac{1}{2},$$
 (2.53)

where $N_{35p}(\text{out})$ is the 35p-state population at the output of the excitation region and N_0 is the total flux of Cs atoms in the ground state 6s at the input of the excitation region.

The mode whereby the saturation conditions are not reached,

$$N_{ns}(\text{out}) \sim N_{35p}(\text{out}) F\sigma(35p \to ns) \frac{1}{\Gamma_{35p} + 1/\tau}, \quad (2.54)$$

where Γ_{35p} is the total radiative decay probability, is optimal for the *n*s level population. The value of this quantity calculated with the aid of FAC [33] is

$$\Gamma_{35p} = 2 \times 10^4 \, \mathrm{s}^{-1} \,, \tag{2.55}$$

and we can assume that

$$\frac{F\sigma(35\mathrm{p}\to n\mathrm{s})}{\Gamma_{35\mathrm{p}}+1/\tau}\simeq\frac{1}{3}\;.$$

In view of (2.53) and (2.54), this gives the flux $N_{ns}(out)$ as

$$N_{ns}(\text{out}) = \frac{1}{3} N_{n_1 p}(\text{out}), \quad \beta_2 = \frac{1}{3},$$
 (2.56)

the laser pump required at the $35p \rightarrow ns$ transition being equal to only

$$F(\omega_{ns \to 35p}) = 0.5 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$$

It is significant that owing to (2.55), the state 35p completely decays on the path $L \sim 50$ cm from the working region to the detector:

$$\exp\left(-\Gamma_{35p}\,\frac{L}{v}\right) = \exp(-20) \ll 1\,.$$

Particular attention should be given to deexcitation of atoms in the *n*s state on their way from the working region to the detector. A calculation of the total radiative decay probability of the states ns, n = 150 for atomic cesium performed by the FAC code [33] yielded the following data:

$$\Gamma_{100s} \simeq 1.1 \times 10^3 \text{ s}^{-1}, \quad \Gamma_{150s} \simeq 0.33 \times 10^3 \text{ s}^{-1}.$$
 (2.57)

Therefore, the flux of Cs atoms in the 150s state that reach the detector at the distance L = 50 cm from the working region $N_{150s}(L)$ is

$$N_{150s}(L) = N_{150s}(\text{out}) \exp(-1.55) = 0.21 N_{150s}(\text{out}),$$

 $\beta_3 = 0.21.$
(2.58)

Simultaneously taking expressions (2.53), (2.56), and (2.58) into account gives the flux of atoms in the *n*s state into the ionization detector:

$$N_{150s}(L) = 0.03N_0. (2.59)$$

Estimates of the photoionization from the *n*s level under irradiation by the laser with $\lambda \sim 0.4 \,\mu\text{m}$, which is employed in the working region for the excitation to the 35p state, show that this process is wholly immaterial at time $\tau = 10^{-4}$ s.

The *ns*-state photoionization due to thermal wall radiation on the way from the excitation region to the detector is negligible. The mixing of the *ns* state with its neighbors $n \pm 1$, $n \pm 2,...$ by thermal radiation extends to only its closest neighbors and is therefore also immaterial (see Section 2.4). This is a characteristic of the experimental scheme under discussion, because the ionization detector records the total flux of excited (Rydberg) atoms without ascribing them to a specific *n*.

The estimates given in this section show that by effecting *ns*-state excitation by an infrared laser with $\lambda \sim 100 \ \mu\text{m}$, it is possible to ensure the acquisition time [compare expressions (2.47), (2.49), (2.60)]

$$T = 10^4 \frac{10^8}{N_{150s}(L)} = \frac{1}{3} \times 10^{14} \frac{1}{N_0}, \qquad (2.60)$$

where N_0 is the initial flux of Cs atoms in the ground state. It follows from (2.60) that for $N_0 = 3 \times 10^8 \text{ s}^{-1}$, the acquisition

time is $T = 10^5$ s. We recall that this acquisition time was obtained under the specific assumptions made above about the values of several characteristic quantities. For instance, the value $\Delta I = 10^{-3}$ Hz of the quantity to be measured corresponds to the spin-polarized xenon density $\sim 3 \times 10^{19}$ cm⁻³, which may turn out to be somewhat overestimated. The transverse velocity in the beam $v_{\perp} \sim 10^{-3} v$ may prove to be underrated. That is why the estimates given in this section require revisions as applied to specific experimental conditions. Furthermore, expression (2.46), which determines the requisite acquisition time, is based on only the Poisson noise in the flux of excited atoms into the detector. The inclusion of additional, excess 'technical' noise may entail some increase in T. True, there is some safety margin in the flux N_0 of Cs atoms. It appears realistic to arrange a flux $N_0 \sim 10^9 - 10^{10} \text{ s}^{-1}$, i.e., a flux about an order of magnitude higher than the flux $N_0 = 3 \times 10^8 \text{ s}^{-1}$ adopted when estimating the time *T*.

The issue of spectral measurements requires special consideration. In this case, account must be taken of the fact that the proposed technique does not involve measurements of the absolute transition frequency with the accuracy $\Delta I = 10^{-4} - 10^{-3}$ Hz, and this 'jitter' should be recorded in the spectral line at the nuclear precession frequency. This requires maintaining the IR-laser line position on the slope of the absorption line of the $n_1 p \rightarrow n_2$ transition with the aid of an active stabilization system (see Fig. 20). Considering that the acquisition time $T = 10^5 - 10^6$ s in the synchronous corresponds to the frequency detection band $\Delta v \approx 2 \times 10^{-6} - 2 \times 10^{-7}$ Hz, a 'technical jitter' of the laser frequency (or the electric field instability reduced to the Stark shift) can be tolerated at the level of a fraction of a kilohertz. This is undoubtedly an intricate task: the precession phase must be recorded, the laser frequency may require locking to an external reference, special means may be needed for stabilizing the electric intensity E_0 , etc.

Summarizing the contents of this section, we conclude that exciting the $n_1p \rightarrow ns$ (n = 150) transition with a SIFIR-50 laser with $\lambda = 118.83 \ \mu m$ or $\lambda = 184.31 \ \mu m$, it is possible to ensure a reasonable acquisition time $T = 10^5 - 10^6$ s required to determine the EDM of atomic 1^{29} Xe accurate to $2 \times 10^{-28} \ e$ cm.

2.6 Spurious instrumental effects

Detailed analyses and discussions of spurious instrumental effects are possible with reference to a specific experimental complex. In what follows, we therefore stay within the framework of the basic experimental scheme and restrict ourselves to the discussion of only a few typical examples of instrumental effects δI_{instr} that require control and suppression if we are to measure the desired effect $\Delta I \propto \mathbf{E}_0 \mathbf{d}$ [see formula (2.34)].

At the locus of an alkali atom, the synchronous precession of the polarization **P** and the magnetization **M** produces both the field \mathbf{E}_{EDM} and the magnetic field \mathbf{H}' of the quasistatic magnetic dipole **M** at the precession frequency. The last of these gives rise to the Zeeman effect. Extracting the Stark shift $\Delta I \propto \mathbf{E}_0 \mathbf{d}$ of the $ns^2 S_{1/2}$ against the background of a much stronger symmetric Zeeman splitting (broadening) appears to be feasible if the external field \mathbf{E}_0 is modulated at the frequency Ω_1 . In this case, the signal at the frequency $\Omega_1 \pm \Omega$ corresponds to the Stark shift $\Delta I \propto \mathbf{E}_0 \mathbf{d}$, while the Zeeman splitting makes a contribution at the frequency Ω .

More hazardous is the effect arising due to the motion of an alkali atom, which is referred to below as the effect of the vector product $\mathbf{v} \times \mathbf{H}'$.

In the frame of reference of an alkali atom traveling with a velocity \mathbf{v} , it experiences the electric field

$$\mathbf{E}' = \frac{1}{c} \, \mathbf{v} \times \mathbf{H}' \,, \tag{2.61}$$

where \mathbf{H}' is the magnetic field of a quasistatic magnetic dipole (see Ref. [30, § 29]),

$$\mathbf{E}' = \frac{1}{c} \frac{4\pi}{3} \left(\frac{R_0}{R}\right)^3 \left\{ \mathbf{v} \times \left[\mathbf{M} - 3(\mathbf{n}\mathbf{M}) \, \mathbf{n} \right] \right\}, \tag{2.62}$$

where $\mathbf{M} = N\mathbf{\mu}$, $\mathbf{\mu}$ is the nuclear magnetic moment of the ¹²⁹Xe atom: $\mathbf{\mu} \sim 10^{-24}$ CGSE, N is the density of polarized xenon atoms, and **n** is the unit vector that originates at the center of the sphere and is directed at the alkali atom.

We make estimates in the case where **n** is perpendicular to the plane (\mathbf{E}_0 , **P**). The desired effect of the proposed method $\Delta I \propto \mathbf{E}_0 \mathbf{E}_{EDM}$ and the spurious effect $\delta I_{instr} \propto \mathbf{E}_0 \mathbf{E}'$, which emerges due to expression (2.62), depend on the precession frequency Ω in a similar manner. Additional modulation of the electric field \mathbf{E}_0 , which enables eliminating some spurious effects, is useless in this case: both $\mathbf{E}_0 \mathbf{E}_{EDM}$ and $\mathbf{E}_0 \mathbf{E}'$ would show up at the same combination frequencies.

Apart from the term ΔI proportional to $\mathbf{E}_0 \mathbf{E}_{\text{EDM}} = (4/3) \pi N \mathbf{d}$ in (2.29), the expression for the Stark shift involves the additional term

$$\delta I_{\text{instr}} \propto \mathbf{E}_0 \mathbf{E}' = \frac{1}{c} \mathbf{E}_0 (\mathbf{v} \times \mathbf{H}') \,.$$
 (2.63)

The experiment should provide for the conditions for minimizing expression (2.63). When the velocity v is aligned with \mathbf{E}_0 (this is precisely the situation provided for in the above description of the basic experimental scheme), \mathbf{E}' in formula (2.61) is perpendicular to \mathbf{E}_0 and the scalar product $\mathbf{E}_0\mathbf{E}'$ is equal to zero. The same statement $\mathbf{E}_0\mathbf{E}' = 0$ is true for other velocities \mathbf{v} as long as they remain in the plane (\mathbf{E}_0 , \mathbf{P} , in expression (2.63), we have

$$\mathbf{E}_0 \mathbf{E}' \sim \frac{v_\perp}{c} E_0 H', \qquad (2.64)$$

where v_{\perp} is the component of the velocity **v** normal to the plane (**E**₀, **P**); the quantity **E**₀**E**' changes sign when v_{\perp} changes sign. This implies that the effect **v** × **H**' can, at least in principle, be suppressed due to averaging over the velocity distribution of alkali atoms in the beam.

Critically important is the fact that the proposed experiment allows reliably monitoring the unaveraged, residual effect of $\mathbf{v} \times \mathbf{H}'$. The point is that a common cell is believed to contain the nuclear-spin-polarized atoms of both ¹²⁹Xe and ³He under perfectly identical conditions. Because the ³He nuclear EDM should be much smaller than the ¹²⁹Xe nuclear EDM, the signal at the ³He nuclear spin precession frequency allows monitoring the result of averaging the quantity δI_{instr} ($\Delta I = 0$ at this frequency).

As noted above, the requirements that the proposed method imposes on the stability and controllability of the magnetic field **B** are significantly slackened. The precession frequency is merely an indicator of the contribution ΔI to the signal and not the main quantity to be measured. As a result, extremely strict requirements imposed on magnetic screens

3. Conclusions

The latest, most advanced experiment in the search for the EDM of ¹²⁹Xe atoms yielded the following upper EDM bound [18]:

$$|d(^{129}\text{Xe})| < 4 \times 10^{-27} e \text{ cm}.$$

Newly planned experiments in search of the ¹²⁹Xe atomic EDM pursue the goal of reaching the level $|d|^{(129}\text{Xe})| \sim 2 \times 10^{-28}e$ cm corresponding to the theoretical predictions of the ¹²⁹Xe atomic EDM, which are now discussed in the literature, anticipated in different versions of the extended standard model (see, e.g., Refs [2, 34]).

Therefore, the goal is to gain important information for the new physics beyond the standard model.

Despite the wide diversity of projects searching for the ¹²⁹Xe atomic EDM known from the literature, their key point involves measuring the precession frequency Ω , Eqn (1.3), in collinear magnetic **B** and electric **E** fields and extracting the shift of this frequency $\Delta\Omega$, Eqn (1.4), under sign reversal of the field **E**.

In this paper, a different method is proposed for measuring the EDM of 129 Xe atoms: it involves recording the electric EDM field of spin-polarized 129 Xe atoms from the Stark shift of the *n*s Rydberg levels of alkali atoms, which is induced by this field.

Until the new experimental method has been realized and comprehensively tested, one cannot unambiguously say that it offers advantages over the conventional method. We may only emphasize its fundamental features that give hope for overcoming the problems encountered in the quest and measurement of the ¹²⁹Xe atomic EDM at the level of $\sim 2 \times 10^{-28} e$ cm.

This method is specific in that the focus of the experiment is transferred to the accuracy of frequency measurements of the Stark shift in the infrared spectral region. The quantity ΔI in (2.34) to be directly measured exceeds the quantity $\Delta \Omega$ measured in the most successful experiment [18] by several orders of magnitude (up to $10^4 - 10^5$). The requirements of the high-precision measurement of the precession frequency Ω and of the stability of the magnetic field **B** are lowered manyfold.

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