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## Role of collisions in the search for an electron electric-dipole moment

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We study the role of atomic collisions in future measurements of an intrinsic electric-dipole moment (EDM) of the electron using laser-cooled Cs atoms in an optical trap or in an atomic fountain. We find that the shift in frequency and the line broadening caused by collisions may eventually limit the achievable sensitivity of these EDM experiments. We present the results of a coupled-channel calculation of these quantities and discuss the symmetry aspects and magnetic-field dependences.

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Measurements of the electric-dipole moment  $d_e$  have become an important tool in the search for new CPviolating interactions outside the standard model [1]. An electron may possess an electric-dipole moment (EDM) only by virtue of interactions that violate time inversion T (equivalent to CP) and space inversion P. Since its CP violation is small, the standard model predicts an extremely small electron EDM  $d_e \sim 10^{-37} e \text{ cm}$  [1], which is orders of magnitude below the current experimental bound of  $1 \times 10^{-26} e \text{ cm}$  [2]. However, extensions of the standard model generally have CP violating interactions which produce a much larger electron EDM, which can be of the order of the current experimental bound [1]. Thus, improvements in the present bound on  $d_e$  are of great importance.

The current experiments use atomic vapors or beams of Cs or Th atoms [2,3]. Due to relativistic effects [4], these heavy atoms acquire an atomic electric-dipole moment  $d_a = Rd_e$ , with  $R(Cs)=114 \pm 3$  and  $R(Tl)=-600 \pm$ 400 [5]. The experiments search for the first-order Stark shift of ground-state Zeeman sublevels that would result from the interaction of an atomic moment  $d_a$  with an applied electric field  $\vec{E}$ . Both systematic and statistical errors limit these experiments. For atomic beam experiments [2], the dominant systematic error is the interaction of the atom's magnetic moment with the motional magnetic field  $\vec{B}_{mot} = \vec{E} \times \vec{v}/c^2$ , where  $\vec{v}$  is the atomic beam velocity. To the extent that  $\vec{B}_{mot}$  has a component along the quantization axis, this interaction gives a shift which is linear in E and mimics an atomic EDM. Cell experiments do not suffer from this error since the average atomic velocity is zero. However, the sensitivity of cell experiments is reduced because the maximum electric field which can be applied is only a few kV/cm, in contrast to the 100 kV/cm which can be applied in a beam. Accurate control of the electric field in a cell can also be difficult.

One promising route to improve the current experiments is to laser cool the atoms [6]. In particular, lasercooled Cs atoms offer several powerful advantages for an electron EDM measurement. A Cs vapor with a density  $n > 10^{10}$  cm<sup>-3</sup> and temperature below 10  $\mu$ K may be readily produced in the laboratory [7]. At these extremely low temperatures, the motional magnetic field errors that can limit atomic beam experiments are greatly reduced. Also, the coherent interaction times can be much greater than in an experiment with room temperature atoms. Linewidths below 1 Hz could be obtained, in contrast to the 50–100-Hz linewidths of the conventional experiments [2,3]. This reduced linewidth should lead to lower statistical errors.

An additional error which can limit EDM measurements arises from uncontrolled changes in the ordinary. second-order Stark effect that results from imperfect control of the electric field [3]. This error has been dramatically reduced in the cesium cell experiments, but would still be very troublesome if the accuracy of the electron EDM were to be improved beyond  $10^{-27} e \text{ cm}$ in a Cs experiment [8]. A further advantage of a lasercooled Cs experiment is that the linewidth can be less than the splitting between neighboring  $\Delta m = 1$  Zeeman resonances that results from the ordinary, second-order Stark effect (about 40 Hz at  $E = 10^5$  V/cm) [9]. Because of this, it would be possible to directly determine the splitting between the m = -1 and m = +1 Zeeman sublevels in either of the two ground-state hyperfine components. This interval could be coherently probed with a two-photon variant of the Ramsey method [10], following optical pumping of the atoms into the m = 0 state. The second-order Stark shift is absent from this interval because the  $m = \pm 1$  sublevels shift by the same amount. Errors due to imperfect control of the electric field would therefore be eliminated from a Cs EDM measurement using this transition.

At the high densities and low temperatures which are favorable for a laser-cooled Cs atom experiment, line shifts and damping of atomic coherences due to collisions between the trapped atoms could become quite significant. Although not in themselves a systematic error, collisional line shifts could limit the accuracy of the experiments if they exhibit random variations due to changes in atomic density. In addition, collisional damping of

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coherences could limit the resolution of the Zeeman resonances. In this paper we present expressions for the collisional frequency shift to the m = -1 to m = +1interval, as well as the corresponding line broadening. We find that an additional advantage of this transition is that its collisional shift vanishes if opposite Zeeman sublevels are occupied with equal partial densities. Finally, we discuss the implications of our results for the proposed experiments. Note that our calculations apply both to EDM experiments using a cold atomic Cs vapor in a trap and those based on an atomic fountain.

Our results closely follow a previous calculation of the collisional shift of the Cs hyperfine "clock" transition [11]. We start from the quantum-mechanical Boltzmann equation for a homogeneous system describing the time evolution of the one-particle density-matrix element  $\rho_{\alpha\beta}$  due to collisions [12]. It can readily be shown that in the situation of interest this equation reduces to

$$\left. \frac{d\rho_{\alpha\beta}}{dt} \right|_{\text{coll}} = n\rho_{\alpha\beta} \sum_{\nu} \rho_{\nu\nu} \sum_{\mu} \left[ (1+\delta_{\alpha\mu})(1+\delta_{\beta\mu})(1+\delta_{\alpha\nu})(1+\delta_{\beta\nu}) \right]^{1/2} \langle v\sigma_{\alpha\beta,\nu\to\mu} \rangle_{\text{therm}}, \tag{1}$$

where  $\alpha$  and  $\beta$  are the hyperfine states probed in the experiment:  $|f,1\rangle$  and  $|f,-1\rangle$  with f equal to either 3 or 4. We label the Cs hyperfine states with the quantum numbers f and  $m_f$ ,  $\vec{f} = \vec{s} + \vec{i}$  being the total spin vector. The atomic density is denoted by n, and  $v = 2\hbar k/m$  is the relative velocity of the Cs atoms. The left-hand side corresponds to the time derivative of the single-particle distribution function in the classical Boltzmann equation, while the righthand side is essentially the product of single-particle distribution functions, which occurs in the Boltzmann collision term. The complex "cross sections"  $\sigma_{\alpha\beta,\nu\to\mu}$  are associated with the contribution of collisions between an atom in a coherent superposition of the  $\alpha$  and  $\beta$  states and a  $\nu$  state atom, the latter thereby relaxing to the  $\mu$  state. These frequency-shift cross sections can be expressed in terms of S-matrix elements:

$$\sigma_{\alpha\beta,\nu\to\mu}(E) = \frac{\pi}{k^2} \sum_{l} (2l+1) [S^l_{\{\alpha\mu\},\{\alpha\nu\}}(E) S^{l*}_{\{\beta\mu\},\{\beta\nu\}}(E) - \delta_{\mu\nu}].$$
(2)

In the following we leave out the inelastic contributions which generally amount to at most 10% of the elastic shifts and widths and therefore do not change the predicted orders of magnitude. From Eq. (1) we find [12,11] expressions for the line shift  $\delta \omega_c$  and broadening  $\Gamma_c$  linear in the partial densities  $n_{\nu}$  (with  $\nu$  including the  $\alpha$  and  $\beta$ states):

$$\delta\omega_c = \sum_{\nu} \sum_{l} n_{\nu} \langle v \lambda_{\nu}^l \rangle_{\text{therm}}, \qquad (3)$$

$$\Gamma_c = \sum_{\nu} \sum_{l} n_{\nu} \langle v \sigma_{\nu}^l \rangle_{\text{therm}}.$$
 (4)

Note that contrary to Ref. [11] we here have incorporated the  $(1 + \delta)$  factors in the  $\lambda$  and  $\sigma$  cross sections.

The singlet and triplet potentials used in the two-atom Hamiltonian are the same as those used earlier in our group to calculate line shifts and broadenings in the Cs atomic fountain [11], except for a modification following from a recent analysis [13] of experimental collisional shifts. In that paper it was shown that the shifts are independent of the detailed form of the inner parts of the potentials, while the outer parts have been sufficiently accurately determined by a thorough analysis [14] of spectroscopic data. The inner parts enter the calculated shifts only in the form of accumulated phases  $\Phi_S$  and  $\Phi_T$  of the radial wave functions, or equivalently in the form of the corresponding values  $v_{DS}$  and  $v_{DT}$  of the vibrational quantum number at the dissociation limit. We choose the values  $v_{DS} = -0.08$ ,  $v_{DT} = -0.04$  found in Ref. [13].

It is of importance to stress that we assume the electricfield strength to be small with respect to the typical values where the S = 0 and S = 1 electronic wave functions, and consequently also the corresponding Born-Oppenheimer potentials, are significantly changed by the external constant electric field. It is to be expected that the order of magnitude of the frequency shifts and line broadenings to be calculated in the following will not be changed until field strengths are reached where they are comparable to the internal atomic field on the valence electron; i.e., values of order  $\mathcal{E}=E_{\rm ion}^2/(4e^3/4\pi\epsilon_0)\approx$  $0.3\times10^8$  V/cm for Cs atoms. With  $\mathcal{E}\lesssim10^5$  V/cm we are far from that regime. An estimate of the *R*-dependent second-order Stark shift on the basis of Sundberg's groupdipole interaction model [15] indeed shows that the dominant contributions quadratic and cubic in the atomic polarizability correspond to negligible interaction terms.

In Figs. 1 and 2 we present the zero field energy dependence of the partial cross sections  $\lambda$  and  $\sigma$  for s-wave scattering in the energy range  $10^{-8}$  to  $10^{-3}$  K for the experiment based on the  $|3, -1\rangle \leftrightarrow |3, 1\rangle$  transition. The contribution for p-wave scattering is only of some importance above approximately  $10^{-4}$  K, and because of Bose symmetry there is even no  $\lambda_{3,1}$  cross section at all. The calculation of the cross sections at zero magnetic field is no restriction since these quantities hardly depend on B for field values aimed at in the experiment, which is about  $10^{-6}$  T at most. However, we will return to this field dependence later. A consequence of the absence of a magnetic field is that  $\lambda_{f,m_f} = -\lambda_{f,-m_f}$  and  $\sigma_{f,mf} = \sigma_{f,-m_f}$ , which follows from a simple symmetry argument. A very important aspect of this relation between  $\lambda_{f,m_f}$  and  $\lambda_{f,-m_f}$  is that the line shift  $\delta \omega_c$  depends on the differences of the partial densities of the  $|f, m_f\rangle$ and  $|f, -m_f\rangle$  states, and can in principle be made arbitrarily small. This is a very favorable circumstance for a precise EDM measurement.

We now turn to the magnetic-field dependence of the



FIG. 1. Line shift cross sections  $\lambda_{f,|m_f|}$  for l = 0 and B = 0, as a function of collision energy for the  $|3, -1\rangle \leftrightarrow |3, 1\rangle$  transition.

 $\lambda_{\nu}$  cross sections. Figure 3 shows this dependence for the  $\lambda_{3,-1}$  and  $\lambda_{3,1}$  cross sections in the case of the  $|4,-1\rangle \leftrightarrow |4,1\rangle$  transition, which implies collision partners with f = 3 and 4. The  $\lambda_{4,m_f}$  in this case show no magnetic-field dependence, which is also the case for the  $\lambda_{3,m_f}$  cross sections when the  $|3,-1\rangle \leftrightarrow |3,1\rangle$  transition is considered. For the explanation we will concentrate on the transition  $|4, -1\rangle \leftrightarrow |4, 1\rangle$ . The  $\lambda_{4,m_f}$  cross sections contain S-matrix elements pertaining to the collision of two atoms that are both from the upper manifold. The total internal energy then only depends on the total magnetic quantum number  $M_F$  and the magnetic-field strength. Because transitions to other  $M_F$  values are prohibited, all the possible channels are either degenerate with the incoming channel, or have an energy which lies one hyperfine splitting lower, too far for their Zeeman splitting to have an appreciable influence on the collision. Changing the magnetic field does not lift this degeneracy, and for the field values considered no opening or closing of channels can occur. However, the  $\lambda_{3,m_f}$  cross sections contain S-matrix elements pertaining to the collision of one atom from the upper, and one from the lower mani-



FIG. 2. Line broadening cross sections  $\sigma_{f,|m_f|}$  for l = 0 and B = 0, as a function of collision energy for the  $|3, -1\rangle \leftrightarrow |3, 1\rangle$  transition.



FIG. 3. Line shift cross sections  $\lambda_{3,-1}$  and  $-\lambda_{3,1}$  as a function of magnetic field at  $10^{-5}$  K for the  $|4,-1\rangle \leftrightarrow |4,1\rangle$  transition.

fold. For this case it can readily be seen that because of the different Landé factors of the two manifolds the total internal energy also depends on the specific  $m_f$  value of the lower manifold atom. This implies a magnetic-field dependence that differs from one channel to another. By changing the field strength the splitting between these channels changes, and opening and closing can occur in this situation. This gives rise to the structure seen in Fig. 3, where indeed the cusps in the  $\lambda$  cross sections can be identified with channel thresholds.

On the basis of these results it is possible to estimate the potential impact of collisions on a laser-cooled Cs atom experiment based on the  $|3, -1\rangle \leftrightarrow |3, 1\rangle$  transition at frequency  $\nu_{-1,1}$ . An experiment might be based either on an atomic fountain [6] or on a blue-detuned far-off resonance optical dipole force trap [16]. In the best case, the measurement accuracy will be limited only by the resonance linewidth and statistical fluctuations due to the finite coherent interaction time T and number of atoms N. Likely conditions in an atomic fountain are T = 0.25 s,  $N = 10^7$  to  $10^{10}$ , a temperature  $E = 2.5 \ \mu \text{K}$ , and an atomic density  $n = 10^7$  to  $10^{10}$ cm<sup>-3</sup>. Likely conditions in a trap are T = 10 s,  $N = 10^5$ to 10<sup>8</sup>,  $E = 50 \ \mu\text{K}$ , and  $n = 10^{9}$  to  $10^{12} \ \text{cm}^{-3}$ . Assuming a Ramsey interrogation of duration T, the resonance linewidth will be  $\Delta \nu_{\rm FWHM} = 1/2T$ , and the shot-noise limit to the frequency resolution for a single Ramsey interrogation will be  $\Delta \nu_{\rm SN} = (2\pi T N^{1/2})^{-1}$  [17]. For the fountain  $\Delta \nu_{\rm FWHM} = 2.0$  Hz and  $\Delta \nu_{\rm SN} = 200$  to 6.3  $\mu$ Hz, whereas for a trap  $\Delta \nu_{\rm FWHM} = 0.05$  Hz and  $\Delta \nu_{\rm SN} = 50$ to 1.6  $\mu$ Hz, for the range of N listed above.

In order to realize these limits, the collisional dephasing rate  $\Gamma_c$  must be less than  $\Delta \nu_{\rm FWHM}$ . This rate changes from  $5 \times 10^{-5} {\rm s}^{-1}$  to  $5 \times 10^{-2} {\rm s}^{-1}$  for the conditions given above for the fountain, and from  $1.2 \times 10^{-3} {\rm s}^{-1}$  to  $1.2 {\rm s}^{-1}$  for the conditions given above for the trap. Thus collisional dephasing of coherence is unlikely to play a significant role in a fountain experiment, but it could limit densities to values well below  $10^{12} {\rm cm}^{-3}$  in a trap experiment if interrogation times much longer than 1 s are to be used. In addition, the collisional frequency shift to  $\nu_{-1,1}$  is  $\Delta \nu_{\text{coll}} = 1.6 \times 10^{-13} [n(\text{cm}^{-3})] (\rho_{3,-1} - \rho_{3,1})$  Hz at  $E = 50 \ \mu\text{K}$ , and 4 times greater at 2.5  $\mu\text{K}$ .

Although this shift is not a systematic shift since it does not change with a reversal of the electric field, it may degrade the resolution if it fluctuates by an amount larger than  $\Delta\nu_{\rm SN}$ . For example, for the trap conditions given above, and a density of  $10^{10}$  cm<sup>-3</sup>, the shift of  $\Delta\nu_{\rm coll} = 1600(\rho_{3,-1} - \rho_{3,1})$  µHz must be held constant to better than 10 µHz. This may be accomplished by keeping  $\rho_{3,-1} - \rho_{3,1}$  balanced to within 0.5%. The choice of the m = -1 to m = 1 transition makes this condition significantly easier to satisfy, since the shift vanishes when the two state populations are equal. The collisional shifts could be problematic for the largest numbers and densities of atoms given above.

At an electric field of  $\mathcal{E} = 10^5$  V/cm, the statistical uncertainty in the electron EDM is given by  $\Delta d_e = 2h\Delta\nu_{\rm SN}/R({\rm Cs})$   $\mathcal{E} = 7.3 \times 10^{-22}\Delta\nu_{\rm SN}({\rm Hz})e\,{\rm cm}$ . Thus if in a single interrogation  $\Delta\nu_{\rm SN} = 10 \ \mu{\rm Hz}$ ,  $\Delta d_e = 7.3 \times 10^{-27}e\,{\rm cm}$ . For either a fountain or trap experiment, extended averaging over many Ramsey interrogation periods could lead to statistical uncertainties below  $10^{-28}e\,{\rm cm}$ , which is an improvement by two orders of magnitude over the current bound. Whether such improvement could be realized depends on whether the shot-noise limit can be realized in practice, and whether systematic errors can also be reduced to these low values. Other effects which must be considered include technical noise, field plate leakage current errors [2,3], magneticfield inhomogeneity and noise, residual motional magnetic field error in a fountain experiment, and ac Stark shifts in a trap experiment. Reduction of all errors to the  $10^{-28}$  level would be a challenging task, and a detailed discussion is beyond the scope of this paper. We note that the ac Stark shift is not a systematic one since it does not change with a reversal of the electric field, and that the Zeeman coherence relaxation time due to photon scattering from the trapping fields could exceed T = 10 s [16]. Also, the cold-atom experiments would have the advantages of very narrow linewidths, the elimination of electric-field reversal errors, and zero or very small motional magnetic-field error. It may be expected that an extension of a previous measurement [18] and analysis [13] of collisional shifts for the clock transition  $|4,0\rangle \leftrightarrow |3,0\rangle$  and future EDM experiments will yield enough information to further restrict the uncertainties in the features of the triplet and singlet potentials relevant for collisions at ultralow energies, such as the scattering lengths.

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