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# Spin-exchange frequency shift in a cesium atomic fountain

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In connection with experiments aiming at the improvement of the cesium atomic beam clock by means of a fountain of laser-cooled cesium atoms, we present expressions for the line shift and line broadening due to collisions between cesium atoms. The coherent collision cross sections occurring in these expressions are calculated by means of the coupled-channel method. The magnitude of the calculated shift implies that spin-exchange collisions are an important factor in limiting the anticipated accuracy of a future cesium fountain clock and of other intended fountain experiments.

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Atomic clocks are based on the observation of the oscillating magnetic moment provided by a coherent pair of hyperfine levels. The performance of such an atomic clock can be improved by lengthening the time interval over which the unperturbed oscillation of the atomic "pendulum" is observed. In the 1950s various possibilities were explored to lengthen this observation time. Ramsey [1] considered the possibility of confining atoms in a storage ring with inhomogeneous magnetic fields or to store atoms in a storage cell with suitably coated walls. An alternative possibility, explored by Zacharias [2], was to use an atomic fountain, i.e., a collection of slow atoms moving in a ballistic orbit and undergoing an rf pulse both on their way up and on their way down with a long time in between. Due to a bad signal-to-noise ratio associated with an insufficient number of slow atoms in a thermal beam this attempt was unsuccessful. A recent experiment by Kasevich *et al.* [3] with Na atoms has shown that the laser-cooling and trapping techniques which are now available create fascinating new possibilities in this regard [4]. Presently, atomic fountain work with laser-cooled Cs atoms is being conducted by Clairon *et al.* [5], Chu [6], and others [7]. Since most of the limiting factors of a conventional clock are considerably reduced, it is anticipated that this development will lead to an unprecedented accuracy: about $10^2$ better than any existing clock [5]. The new atomic fountains also hold great promise for other applications in which the ultraprecise measurement of phase shifts is involved, such as the search for a possible atomic electric dipole moment due to parity and time reversal violation, quantum-nondemolition (QND) measurements of small photon numbers, and atomic interferometry. As we will show, a fountain experiment may also shed light on the possibility of attaining Bose condensation in a Cs trap.

To improve the signal-to-noise ratio atomic densities in atomic fountains are being increased to the point where the line shift due to interatomic collisions between the cold cesium atoms might become significant and even spoil the above-mentioned expectations about the spectacular improvement of the cesium clock and other applications to be achieved with a fountain. In this paper we calculate this crucial quantity, which is the only frequency shift that cannot be reduced to lowering the atomic velocity and which seems to be the main missing piece of information about the atomic cesium fountain [6]. Previous calculations [4] of the spin-exchange shift for the conventional cesium clock have neglected the atomic hyperfine splitting which is a bad approximation for the microkelvin temperatures prevailing in a cesium fountain, as we will show presently. In the following we present expressions for the collisional frequency shift, as well as for the corresponding line broadening. In addition, we calculate the coherent cross sections occurring as coefficients in these expressions and study their dependence on collision energy, orbital angular momentum, and occupations of hyperfine levels. Finally, we discuss the implications for present and future cesium fountains.

A calculation of line shift and broadening may be based conveniently on the quantum Boltzmann equation [8,9]. This leads to the equation

$$\left.\frac{d}{dt}\rho_{\alpha\beta}\right|_c = \rho_{\alpha\beta}\sum_\nu n_\nu \sum_\mu [(1+\delta_{\alpha\mu})(1+\delta_{\beta\mu})(1+\delta_{\alpha\nu}) \times (1+\delta_{\beta\nu})]^{1/2} \langle v\sigma_{\alpha\beta,\nu\rightarrow\mu}\rangle \equiv -(\Gamma_c - i\delta\omega_c)\rho_{\alpha\beta}$$

for the collisional time evolution of the nondiagonal density-matrix element $\rho_{\alpha\beta}$ describing the coherence of the $\alpha=|3,0\rangle$ and $\beta=|4,0\rangle$ hyperfine states $|F,M_F\rangle$. The collisional shift $\delta\omega_c$ and broadening $\Gamma_c$ thus depend linearly on the partial hyperfine densities $n_\nu$. The complex coefficients $\langle v\sigma\rangle$ are associated with the contribution of collisions in which a $\nu$ state atom makes a transition to a $\mu$ state in colliding with an atom which is in a superposition of the $\alpha$ and $\beta$ states. The brackets $\langle\,\rangle$ stand for an average over the distribution of relative velocities $v=2\hbar k/m$ and corresponding energies $E$ pertaining to the experimental circumstances in the fountain. The complex "cross sections" are crossterms of interfering $S$-matrix elements:

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

the curly bracket notation denoting symmetrized (even orbital angular momentum $l$) and antisymmetrized (odd $l$) two-atom hyperfine states.

 

The $S$-matrix elements have been calculated using an effective two-atom Hamiltonian [10], comprising atomic hyperfine interactions and singlet or triplet potentials. The singlet Cs-Cs potential $V_0$ is taken from work by Weickenmeier *et al.* [11], where it is derived from precise and extensive measurements of $Cs_2$ rotation-vibration energies. No comparable measurements are as yet available for the triplet potential $V_1$. Its tail beyond $r_1 = 15.6a_0$ can, however, be reliably determined from the singlet potential by reversing the exchange contribution found in Ref. [11]. For smaller radii the potential has been taken from an *ab initio* calculation by Krauss and Stevens [12]. The parameters $R_e$, $D_e$, and $\omega_e$ following from this calculation agree closely with values estimated in Ref. [11].

The Schrödinger equation for the collision was solved in the form of a set of up to 16 coupled-channel equations in the hyperfine basis for each separate combination of $l$ and total spin magnetic quantum number. The complicated spin structure of the problem was dealt with by means of computer algebra. Writing out the complex cross sections in real and imaginary partial-wave contributions,

$$i\delta\omega_c - \Gamma_c = \sum_{\nu,l} n_\nu (1 + \delta_{\alpha\nu} + \delta_{\beta\nu}) \langle v(i\lambda_\nu^l - \sigma_\nu^l) \rangle ,$$

we present in Fig. 1 the zero-field energy dependence of the partial cross sections $\lambda$ for $l=0$ in the energy range from $10^{-8}$ to $10^{-4}$ K, in Fig. 2 that of the $\sigma$ quantities for $l=0$, and in Fig. 3 that of the $\lambda$ quantities for $l=1$. In the absence of a magnetic field the cross sections are independent of the sign of $M_F$. Because of Bose symmetry odd-$l$ values do not contribute to the $|3,0\rangle$ and $|4,0\rangle$ cross sections. Despite the microkelvin temperatures of the laser cooled atoms, collision energies are clearly not low enough to assume $s$-wave scattering. The $l=1$ partial wave also yields a significant contribution. Note also that the low-energy behavior of all $\lambda_\nu$ and $\sigma_\nu$ except $\sigma_{3,0}$ is proportional

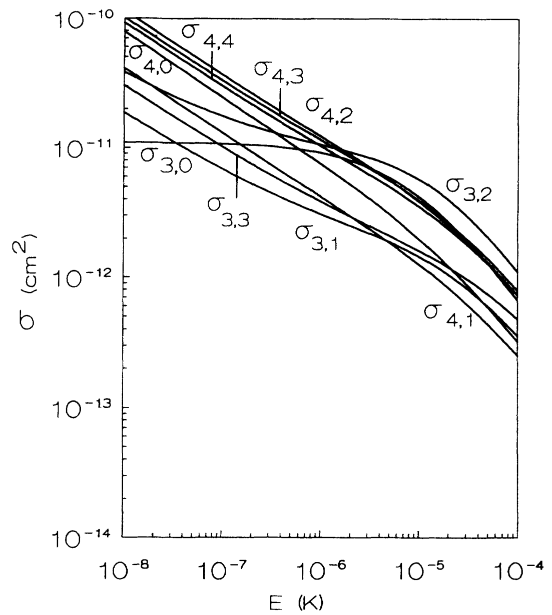


FIG. 2. Line broadening cross sections $\sigma_{F,|M_F|}^l$ for $l=0$ and $B=0$.

to $E^{l-1/2}$, in accordance with the Wigner threshold law [13] for $S$-matrix elements. The exceptional behavior of $\sigma_{3,0} \sim E^{2l}$ can be understood on the basis of unitarity and the spin structure of the symmetric spin state $\{\alpha\beta\}$ with $\alpha = |3,0\rangle$ and $\beta = |4,0\rangle$, which is purely triplet. Some of the $\lambda_\nu$ and $\sigma_\nu$ show an appreciable influence of the bias magnetic field applied to separate the $|3,0\rangle \rightarrow |4,0\rangle$ clock transition from the remaining hyperfine transitions. For the larger partial cross sections, however, including $\lambda_{3,0}$ and $\lambda_{4,0}$, the influence is negligible for field strengths to $2\times10^{-5}$ T. Therefore we do not take the bias field into

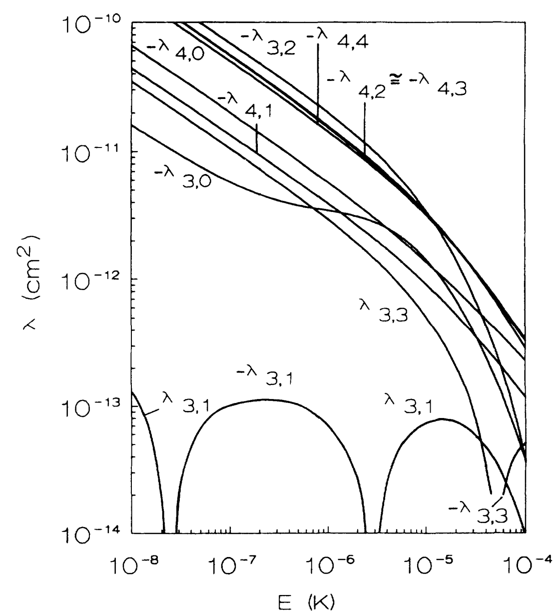


FIG. 1. Line shift cross sections $\lambda_{F,|M_F|}^l$ for $l=0$ and $B=0$, as a function of collision energy.

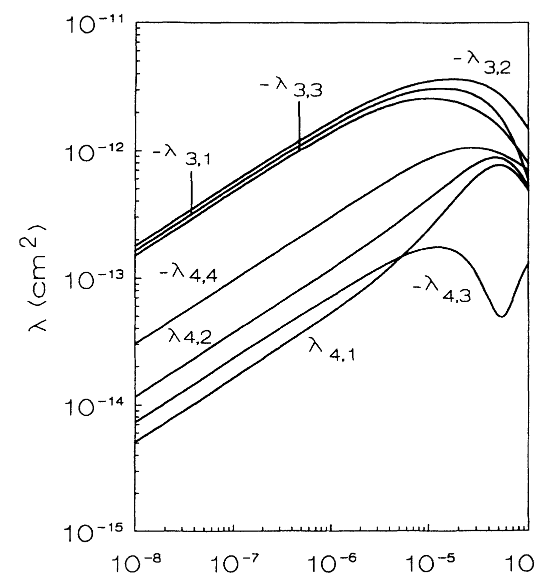


FIG. 3. Line shift cross sections $\lambda_{F,|M_F|}^l$ for $l=1$ and $B=0$.

account in the further discussion. In connection with previous calculations [4] of the collisional line shift for the conventional cesium clock, which neglected the hyperfine splitting, we stress that this approximation leads to vanishing $\lambda$ values, for all $M_F \neq 0$ states. Figure 1 shows that this is a bad approximation, as mentioned previously.

For a cesium fountain frequency standard the collisional line broadening of order $5\times10^{-11}$ sec$^{-1}$ $n$/cm$^{-3}$ predicted on the basis of our calculations is unimportant, since it is small relative to the inverse transit time except at the highest expected densities. The collisional line shift can be calculated by modeling the time evolution of the partial densities of hyperfine levels and that of $\rho_{\alpha\beta}$ in the specific experimental circumstances. From our calculation it is possible, however, to indicate an order of magnitude. Assuming a uniform distribution over magnetic substates the relative frequency shift is estimated to be of order $1\times10^{-22}$ $n$/cm$^{-3}$. The same result is obtained if the atoms are optically pumped into the $M_F=0$ hyperfine states, so that this will not reduce the frequency shift. In view of the anticipated accuracy [5] ($10^{-16}$) of a cesium fountain clock, we conclude that the spin-exchange frequency shift is a crucial effect to be taken into account at the present and expected densities [5,6] ($10^7$–$10^{10}$ cm$^{-3}$). From the very different values of the $\lambda$ cross sections we also conclude that it would be helpful to reduce the partial densities of the substrates with the largest $\lambda$ values. Furthermore, it does not help to further reduce the temperature: The $E^{-1/2}$ dependence of $\lambda_\nu$ is compensated for by the collision velocity, so that for $l=0$ we calculate essentially the zero-temperature limit.

It is of interest to point out that some uncertainty is associated with the results of the present calculation in connection with the triplet potential for $r<r_1$. The fact that the above-mentioned parameters of the *ab initio* potential used agree closely with values following from Ref. [11] gives some confidence in the reliability of the above results, at least with respect to order of magnitude. Due to the fact that the uncertainty in $V_1$ is restricted to an $r$ interval where the hyperfine splitting is very small compared to the exchange interaction, the quantities $\lambda_\nu$ and $\sigma_\nu$ do not depend on the detailed shape of $V_1$ for $r<r_1$, but only on a single parameter, i.e., the logarithmic derivative of the triplet radial wave function at $r_1$. This makes it easy to get an impression of the range of values which the cross sections can take in principle when the triplet potential for $r<r_1$ is varied over a range of shapes much larger than the uncertainty which is suggested by the above close agreement of potential parameters. Varying the well depth $D_e$ with respect to that of Krauss and Stevens we indeed find a periodic behavior of coupled-channel cross sections and a confirmation of the above-mentioned order of magnitude. From the point of view of the atomic cesium fountain it is clear that spectroscopic measurements of rotationally resolved triplet spectra of the type of Ref. [11] are highly desirable. On the other hand, it is also clear that a single measured collisional frequency shift would in principle be sufficient to determine the above-mentioned logarithmic derivative. Knowledge of the theoretical $\lambda$ quantities would make it possible to correct the experimentally observed cesium frequency for the spin-exchange shift. In this connection it would be crucial to know sufficiently accurately the total and partial densities during and in between the rf pulses. Assuming the relative uncertainties to be of order 10%, which is already rather difficult to achieve [6], the remaining inaccuracy ($\delta\nu/\nu \simeq 1\times10^{-23}$ $n$/cm$^{-3}$) would still be a serious threat to the expected accuracy of cesium fountain clocks. In any case it is a crucial effect to be taken into account in all cesium fountain experiments concentrating on phase shifts between hyperfine states.

The measurement of a single collision shift in a fountain may also be relevant outside the field of atomic fountains. The $\lambda$ quantities depend essentially on the difference $a_S - a_T$ of singlet and triplet scattering lengths. Since the value of $a_S$ is known with sufficient accuracy, a forthcoming fountain experiment may very well give the first reliable information on $a_T$ and thus on the possibility of realizing Bose condensation in a Cs trap for which a positive sign of the scattering length is an unavoidable condition [14].

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