

Eliminating cold-collision frequency shifts

Kurt Gibble

Department of Physics, Yale University, New Haven, Connecticut 06520-8120

Boudewijn J. Verhaar

Department of Physics, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

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A technique to cancel the frequency shift due to cold collisions of laser-cooled atoms is proposed. Results for future clocks based on ^{135}Cs and ^{137}Cs are presented. We find a ^{137}Cs fountain clock can be operated with no cold-collision shift by adjusting the population ratios in the fountain, and it may also be possible for ^{135}Cs . This technique can also be applied to other phase-sensitive measurements using cold atoms, including atom interferometers.

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The best-known application of laser cooling [1] is the potentially dramatic improvement of the accuracy of laser and microwave spectroscopy, especially for future atomic clocks. However, for laser-cooled clocks elastic collisions give rise to a frequency shift that appears to be devastatingly large at the achievable μK temperatures [2,3]. The frequency shift cross section is often larger than the usual elastic velocity-changing cross section, since the frequency shift cross section scales as $T^{-1/2}$ for low temperatures, whereas the velocity-changing cross section is constant. Here we propose a solution to this problem that may allow for the previously expected 100- to 1000-fold improvement in the accuracies and stabilities of atomic clocks.

The next generation of atomic clocks is likely to be based on a microwave transition in a laser-cooled atomic fountain [3,4]. Atomic fountains have been created by collecting Cs atoms in a magneto-optic trap and then cooling them to μK temperatures ($v \approx 2$ cm/s) in a reference frame moving upward at ≈ 2 –5 m/s. The laser beams are then turned off and the atoms travel ballistically up and down under the influence of gravity, returning to the launch region after 0.3–1 s. In a fountain clock, the atoms pass through a microwave cavity on the way up, are turned around by gravity, and then pass through the same cavity a second time. In each pass through the microwave cavity, the atoms experience a $\pi/2$ pulse—the first pulse creates an atomic coherence and the second “converts” the phase difference ϕ between the atomic coherence and the microwave field to a population difference that varies as $\cos(\phi)$.

In the 0.3–1-s interrogation time between microwave pulses, collisions occur. Because the atoms are cold, the collisions are intrinsically quantum mechanical, since the atomic de Broglie wavelengths $\lambda_{\text{dB}} \approx 3000$ Å are much longer than the range of the interatomic potential [5]. These collisions in general shift the phase of the atomic coherence created by the first microwave pulse and produce an apparent frequency shift of the clock. Gibble and Chu measured this shift to be $\delta\nu = -16$ mHz at a cold Cs density of 10^9 cm $^{-3}$, which is a fractional frequency shift of $\delta\nu/\nu = -1.7 \times 10^{-12}$ ($\nu = 9\,192\,631\,770$). For comparison, the accuracy of room-temperature clocks is $\delta\nu/\nu \approx \pm 3 \times 10^{-14}$ [6] and the sum of all other systematics in a laser-cooled fountain

clock should be $\approx 10^4$ times smaller than the cold-collision shift [7]. While a laser-cooled fountain clock operating with reduced cold Cs density currently offers a useful improvement in accuracy and short-term stability over room-temperature Cs clocks, the dominant systematic is the cold-collision frequency shift. If a clock were able to operate at the Cs densities achievable with laser cooling without a frequency shift due to cold collisions, significantly better accuracy and short-term stabilities should be possible.

The measured frequency shift cross sections for ^{133}Cs are $\approx 1/4$ of the maximum possible cross section $\lambda_{\text{dB}}^2/2\pi$. An analysis of the measured shifts showed that the reason for the large s -wave cross section is the proximity of both triplet and singlet bound states near $E=0$ [8]. Therefore, it appears that ^{133}Cs , which is the basis for the SI (Système International) definition of the second, is an unfortunately poor choice for laser-cooled clocks. Here we propose and analyze the prospects for several other candidates where the cold-collision frequency shift can potentially be eliminated.

The optimal clock candidate should retain many of the features of ^{133}Cs . These include a high-frequency hyperfine transition, a large mass, a good laser-cooling transition, a reasonably high vapor pressure, and foremost, a small cold-collision frequency shift. To achieve a high-frequency stability in a fountain clock, a large and dense cloud of atoms is needed, which suggests alkali atoms, given their excellent laser-cooling properties.

At present, the best clock candidates are ^{87}Rb or another Cs isotope. The ground-state hyperfine transition of ^{87}Rb is 6.834 GHz, which is 25% lower than the frequency of the ^{133}Cs transition. All of the other Rb isotopes have significantly smaller hyperfine splittings or lifetimes of less than one day. Unfortunately, the vibrational energy spectrum near $E=0$ is not known for Rb, so that the frequency shifts cannot yet be predicted. An analysis of ^{85}Rb photoassociation has already given a prediction for the ^{87}Rb triplet scattering length [9]. Currently, more work is needed to determine the relevant cold-collision properties [10]. While the mass and hyperfine splitting are less than those for the Cs isotopes, the cold-collision shift is the dominant problem and ^{87}Rb has the advantage that it is not considered to be radioactive.

For the Cs isotopes, the interatomic potentials are constrained by the measured frequency shifts for ^{133}Cs . The iso-

topes ^{135}Cs and ^{137}Cs have lifetimes of 3×10^6 and 30 years, and have slightly larger hyperfine splittings of 9.724 and 10.116 GHz versus 9.192 GHz for ^{133}Cs . To show that the cold frequency shifts will be grossly different, we calculate the energy of the last bound or first unbound vibrational state for the singlet and triplet potentials. For the $^{133}\text{Cs}_2$ singlet potential, the fractional number of bound states is $v_{DS}^{133} = 154 - 0.08$ [8,11]. Since the mass of ^{135}Cs is larger, its de Broglie wavelength is shorter, and, in the WKB approximation, $v_{DS}^{135} = (135/133)^{1/2} \times v_{DS}^{133}$. For the triplet potential and likewise for ^{137}Cs , we get

$$\begin{aligned} v_{DS}^{133} &= 154 - \mathbf{0.08}(0.05), & v_{DT}^{133} &= 51 - \mathbf{0.04} \begin{pmatrix} +0.015 \\ -0.06 \end{pmatrix}, \\ v_{DS}^{135} &= 155 + \mathbf{0.07}(0.05), & v_{DT}^{135} &= 51 + \mathbf{0.34} \begin{pmatrix} +0.03 \\ -0.06 \end{pmatrix}, \\ v_{DS}^{137} &= 156 + \mathbf{0.22}(0.05), & v_{DT}^{137} &= 52 - \mathbf{0.28} \begin{pmatrix} +0.05 \\ -0.07 \end{pmatrix}. \end{aligned} \quad (1)$$

Note that the bold nonintegral vibrational quantum number above almost entirely determines all of the cold-collision properties.

While the above results suggest that the frequency shifts in ^{135}Cs and ^{137}Cs may be smaller, an additional technique is needed to eliminate the cold collision shift. We recall that the fountain atoms can be prepared in the $m_F=0$ state, as demonstrated in Ref. [3]. After the first $\pi/2$ pulse, there are atoms in both the $F=4, m_F=0$ and the $F=3, m_F=0$ states. As described in Tiesinga *et al.* [2], the atomic populations in the $F=3$ and $F=4$ states each produce frequency shifts that are generally different. Ideally, the frequency shifts these states produce will be small and have opposite signs, so that, by varying the microwave amplitude for the first pulse, the relative populations of the $F=3$ and $F=4, m_F=0$ states can be adjusted to completely eliminate the frequency shift due to the cold collisions. (This does not work for ^{133}Cs since both states produce nearly the same frequency shift [8].)

The results (1) show that the cold frequency shifts should be different for ^{135}Cs and ^{137}Cs . Unfortunately, the cold shift is not a simple function of v_{DS} and v_{DT} , and a full coupled-channels calculation is needed. In Fig. 1, the results of full coupled-channels calculations, similar to those in Refs. [2] and [8], are shown for ^{137}Cs . In Fig. 1(a), the contour is shown for the frequency shift due to the $F=3, m_F=0$ state in mHz for a density of 10^9 cm^{-3} and a launch temperature of $2.5 \mu\text{K}$. In Fig. 1(b), the shift due to the $F=4, m_F=0$ state is shown and Fig. 1(c) shows the ratio of these two frequency shifts. The likely v_{DS} and v_{DT} range we determined from the previously measured shifts is also shown on this graph. Over the likely area in the v_{DS}, v_{DT} plane, the ratio of the two shifts is between $-3:1$ and $-14:1$.

Eliminating the cold shift for a frequency shift ratio of $-10:1$ implies first driving a $\pi/5$ pulse so that the $F=4, m_F=0$ population is $10/11$ and the $F=3, m_F=0$ is $1/11$ in the fountain. Here, the Ramsey fringe contrast is maximized if the second pulse is a $\pi/2$ pulse, and it is straightforward to show that the contrast, as a fraction of that for two $\pi/2$ pulses, is $2p^{1/2}/(1+p)$, where p is the population ratio [12]. Hence, for large p , this scales as $2p^{-1/2}$. Therefore, the contrast, and hence the frequency discrimination sensitivity, is reduced to 60% when the population ratio is 10:1. The $p^{-1/2}$ dependence occurs since the final signal is proportional

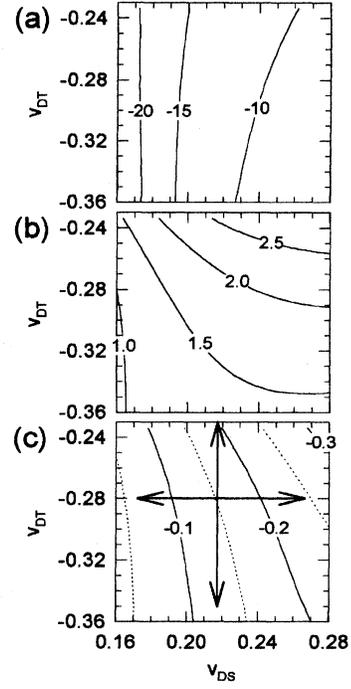


FIG. 1. Calculated frequency shifts for a ^{137}Cs fountain in mHz for an average density of 10^9 cm^{-3} and a launch temperature of $2.5 \mu\text{K}$. (a) Frequency shift caused by the $F=3, m_F=0$ state versus v_{DS} and v_{DT} , the nonintegral vibrational quantum numbers at dissociation for the singlet and triplet potentials. (b) Shifts caused by the $F=4, m_F=0$ state. (c) The ratio of the shifts in (a) and (b). The arrows denote the likely ranges of v_{DS} and v_{DT} for ^{137}Cs .

to the atom's coherence amplitude after the first pulse and not the state population. Therefore, cold frequency shift ratios as large as $-14:1$ can be eliminated without losing more than 50% of the maximum frequency discrimination sensitivity [13].

In Ref. [3] it was shown that an $n \rightarrow 0$ density extrapolation of the frequency of a ^{133}Cs fountain may be possible to within 1% of the cold-collision shift. Given the large optical thickness in clocks capable of the highest short-term stabilities, it will be challenging to be certain of density ratios at the requisite 0.5% level. Here, by first eliminating the cold-collision shift, the $n \rightarrow 0$ density extrapolation can be more certain since the population ratio (i.e., microwave field amplitude) will be adjusted so that there is no frequency shift when the density is changed. Therefore, precise density measurements are not needed.

In Fig. 2, the results of calculations for ^{135}Cs are shown. Again, the $F=4, m_F=0$ shift in Fig. 2(b) is small and insensitive to small changes in v_{DS} and v_{DT} . Due to the proximity of a singlet bound state to $E=0$, the $F=3, m_F=0$ frequency shift has a dispersive resonance structure in v_{DS} . This strong variation of the $F=3, m_F=0$ shift and our current knowledge of the potentials does not allow a significant prediction of the $F=3$ shift and therefore any certainty about the clock prospects for ^{135}Cs ; the region where the frequency shift ratio is between $-1:1$ and $-10:1$ constitutes about 15% of the likely area.

Eliminating the cold-collision shift with these quite gen-

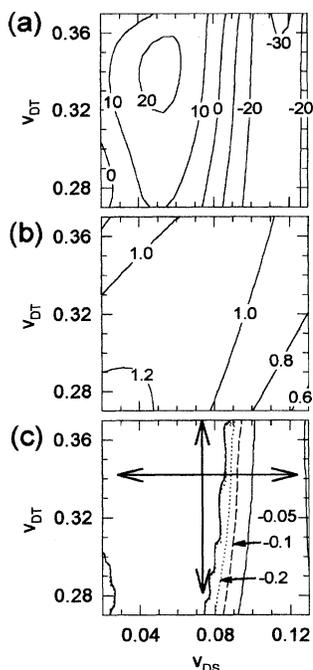


FIG. 2. Calculated frequency shifts for a ^{135}Cs fountain in mHz for an average density of 10^9 cm^{-3} and a launch temperature of 2.6 μK . (a) Frequency shift caused by the $F=3, m_F=0$ state versus v_{DS} and v_{DT} . (b) Shifts caused by the $F=4, m_F=0$ state. (c) The ratio of the shifts in (a) and (b). The arrows denote the likely ranges of v_{DS} and v_{DT} for ^{135}Cs .

eral techniques will allow unprecedented stability and accuracy of atomic clocks. While the low energy β 's of ^{135}Cs create minimal radioactivity concerns, its prospects are un-

certain. On the other hand, the calculations for ^{137}Cs predict superior clock performance, although the clock will have to be designed to use minimal amounts of ^{137}Cs or be designed with proper radiation shielding. For these reasons, it may turn out that ^{87}Rb will be the atom of choice, despite its smaller hyperfine splitting and lighter mass if the unknown cold collision shifts are small or cancellable.

More measurements of ^{133}Cs cold-collision properties or measurements of the cold-collision shifts in ^{135}Cs or ^{137}Cs should precisely test the cold-collision theory. This is important since experimental results from H masers [14,15] are not consistent with calculations [16].

Finally, we note that the cold-collision shift could also be eliminated by choosing a fermionic atom. For two identical fermions in the same spin state, there can be no s -wave scattering, and the p -wave frequency shift goes to zero as T for temperatures $T < 30 \mu\text{K}$. One candidate here is ^{134}Cs , which has a 10.474-GHz transition frequency. Such a clock would operate on an $F=7/2, m_F=-1/2$ to $F=9/2, m_F=1/2$ transition at a bias magnetic field of 3.3 G, where the transition is independent of magnetic field to first order [17]. Unfortunately ^{134}Cs has a half life of only two years and its radioactivity is even more challenging than ^{137}Cs . Another fermion candidate is metastable ^{129}Xe , which has a 5.9-GHz hyperfine transition. A trap has already been demonstrated [18], although higher loading rates and more trapped atoms would be useful.

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