Ultrahigh-resolution two-photon optical Ramsey spectroscopy of an atomic fountain

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We present a semiclassical analysis of ultrahigh-resolution two-photon optical Ramsey spectroscopy of cold neutral atoms falling freely in a fountain. Considering atoms which interact with the same standing-wave laser field twice on their parabolic trajectories and averaging over a broad atomic velocity distribution, we predict a nearly Lorentzian line shape whose width is just the natural linewidth. We have investigated a number of systematic corrections to the atomic resonance frequency, including first-order and second-order Doppler shifts, gravitational red shifts, and ac Stark shifts. First-order Doppler shifts due to vertical motion cancel even if the counterpropagating beams are slightly misaligned, and resolutions below 1 Hz appear feasible with a fountain of modest dimensions.

I. INTRODUCTION

In this paper we explore Doppler-free two-photon optical Ramsey spectroscopy of cold neutral atoms falling freely due to gravity. Using a semiclassical model, we consider a fountain of atoms which interact with the same standing-wave laser field twice on their parabolic trajectories. After averaging over a broad atomic velocity distribution, we predict a simple nearly Lorentzian interference signal whose width is just the natural linewidth. Resolutions below 1 Hz appear feasible with a fountain reaching only a few centimeters above the laser beams. Residual first-order Doppler shifts due to vertical motion cancel even if the counterpropagating beams are slightly misaligned. Optical Ramsey spectroscopy of an atomic beam[1,2] suffers from uncontrolled line shifts caused by phase differences between the two interaction regions. The fountain geometry avoids this problem because the same light field is used for both interactions. A brief summary of the results derived here has already appeared elsewhere.[3]

One of the motivations for this analysis has been the recent progress in high-resolution continuous-wave (cw) two-photon spectroscopy of atomic hydrogen.[4] The hydrogen $1S$-$2S$ transition at 243 nm is of particular interest for precision measurements of fundamental constants and for tests of fundamental physics laws, and its 1-Hz linewidth presents a very tantalizing goal for ultrahigh-resolution spectroscopy. Other atoms with two-photon transitions of similarly narrow widths have been recognized as interesting candidates for optical frequency standards.[5]

To obtain resolutions near 1 Hz, the effective observation time must extend over a good fraction of a second. Unfortunately, such long observation times are not easily achieved for neutral atoms, despite recent spectacular advances in laser radiation pressure cooling.[6–9] Even at a temperature of 1 mK, atoms of an atomic mass M (in amu) are still traveling with a most probable speed of $4/\sqrt{M}$ m/s. Adequate interaction times can be reached in principle by trapping of laser-cooled atoms[9,10] but then the atomic energy levels are perturbed by the strong external electromagnetic trapping fields, and the transition frequency may be shifted by an uncontrollable amount.

Here we show that natural-linewidth-limited spectroscopy with extreme resolution is possible if the Earth's gravitational field is used to further slow laser-cooled atoms so that each projectile can be observed for an extended period near the turning point of its parabolic trajectory. The freely falling atoms can remain completely unperturbed by any external fields except for the two brief passages through the exciting laser field. The basic idea of using gravity to slow atoms is not new.[11,12] More than 30 years ago, Zacharias at MIT built an apparatus 9 m tall for microwave spectroscopy of an atomic cesium fountain. That experiment was not successful because collisions in the orifice of the hot cesium oven eliminated the slow tail of the Maxwellian velocity distribution. This obstacle can be overcome by laser cooling, which provides such low temperatures that even atoms near the peak of the velocity distribution can be slowed by gravity to a standstill over modest distances. Fountain experiments have recently been suggested as a means to study the $1S$-$2S$ two-photon transition in atomic hydrogen.[4,13] However, we do not know of any quantitative analysis of two-photon optical Ramsey spectroscopy in an atomic fountain.

In our model we consider the experimental arrangement shown in Fig. 1. Atoms from a cold source region (or "nozzle"), prepared in the ground state, move upward against the force of gravity towards a single excitation region of two horizontal, nearly counterpropagating laser beams with the same frequency. We are interested in those atoms which pass through this laser field twice as they move along their parabolic trajectories. Any excited atoms falling from above are monitored by a suitable detector positioned just below the light field. As in optical Ramsey spectroscopy of an atomic beam,[1,2] the first passage leaves an atom in a coherent superposition of ground state and excited state. The effect of the second passage then depends on the relative phase of the atom-

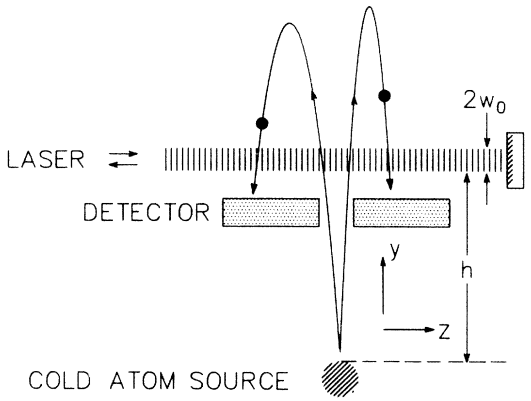

FIG. 1. Scheme for two-photon optical Ramsey spectroscopy of atoms falling freely in a fountain.

field system after the time of flight T between the interactions, giving rise to Ramsey interference fringes in the excitation probability.

In ordinary Ramsey spectroscopy, the atoms travel with constant velocity v between two distinct interaction regions separated by a distance L, so that $T=L/v$. Hence in that arrangement the narrowest fringes are provided by the slowest atoms. In the fountain, however, atoms which enter the light field with vertical speed v spend a time $T=2v/g$ between the interactions, so in this geometry it is the *fastest* atoms which give rise to the fringes with the highest resolution. But the fringe contrast becomes very small if the atoms are *too* fast: the effect of the atoms' first interaction is diminished because of spontaneous decay before they return. (Since it is the interference of the second interaction with the first that causes the fringes, only atoms which are still in a coherent superposition of states when they reenter the excitation region can contribute to the signal.) On the other hand, atoms which are too slow do not reach sufficient height to cross the beams. As a result, only atoms within a relatively narrow range of vertical velocities contribute to the useful interference signal, and the distribution of atomic speeds outside this range is unimportant for the velocity-averaged signal line shape.

In Sec. II, we apply the semiclassical formalism developed in the Appendixes to derive the excitation probability for a single atom in free fall which passes twice through an interaction region of two counterpropagating laser beams. We consider corrections due to beam misalignment, first-order and second-order Doppler shifts, gravitational red shifts, and ac Stark shifts. In Sec. III we determine the velocity distribution of the atoms which reach the excitation region in terms of the velocity distribution at the source. We then average the single-atom excitation probability over the atomic velocity distribution. We point out that, as long as the damping is large enough, the useful velocity range will be narrow compared with the width of the distribution, and an average over all atoms yields a nearly Lorentzian signal, regardless of the exact shape of the distribution. For a Maxwellian velocity distribution at the source, we derive the higher-order corrections to this line shape. Finally, in Sec. IV, we apply our analysis to a particular example, the $1S$-$2S$ transition in atomic hydrogen.

In the Appendixes we briefly develop the theory of two-photon transitions with an eye towards our particular application. The resonant interaction between intense laser beams and atoms is treated with a semiclassical model, introducing vacuum field-atom interactions with appropriate phenomenological damping factors. In Appendix A we derive the semiclassical optical Bloch equations for a general two-level atom, including time-dependent frequency corrections such as ac Stark shifts. We then write down a simple integral expression for the time-dependent excitation probability. In Appendix B, we use conventional techniques to develop an effective semiclassical Hamiltonian for resonant two-photon transitions in the electric dipole approximation (EDA) and the rotating-wave approximation (RWA). We contrast our result with that of quantum electrodynamics, and we discuss the limitations of the usual photon number state treatment. Finally, we explicitly define the circumstances under which our effective Hamiltonian reduces to the form commonly found in the literature.

II. THE SINGLE-ATOM EXCITATION PROBABILITY

In our semiclassical model we assume that the (quantum-mechanical) atoms follow classical trajectories through a classical electromagnetic field. We label the intensities of the two nearly counterpropagating cw laser beams I_1 and I_2, the phases ϕ_1 and ϕ_2, and the propagation vectors $\mathbf{k}_1=k\hat{\mathbf{z}}$ and $\mathbf{k}_2=-k\hat{\mathbf{z}}+\Delta\mathbf{k}$, where $kc=2\pi c/\lambda=\omega$ is the frequency of each laser and $|\Delta\mathbf{k}|^2=2k\hat{\mathbf{z}}\cdot\Delta\mathbf{k}\ll k^2$ allows for a small misalignment. The intensity profile of the light field (e.g., Gaussian) will determine the size and shape of the relatively broad envelope of the Ramsey fringes corresponding to a particular velocity class.[14] However, the field profile has little effect on the central interference signal after averaging over the atomic velocity distribution, so for simplicity we approximate the geometry of the electric fields by rectangle functions of dimension $2w_0$ in the y direction. At $t=0$ an atom which is initially in the ground state enters the light field at $\mathbf{r}(0)=0$ with a nearly vertical velocity $\mathbf{v}(0)=v_x\hat{\mathbf{x}}+v_y(0)\hat{\mathbf{y}}+v_z\hat{\mathbf{z}}$. Under the influence of the Earth's gravitational field $\mathbf{g}=-g\hat{\mathbf{y}}$, the atom exits the interaction region after a time

$$\tau=\frac{1}{g}\{v_y(0)-[v_y^2(0)-4gw_0]^{1/2}\}\approx 2w_0/v_y(0)\ .$$

It then continues on a parabolic trajectory and reenters the light field at the time $T=2v_y(0)/g-\tau$. Finally, at time $t=T+\tau$ the atom leaves the interaction region, travels a short distance, and reaches a metastable detector. (Throughout this analysis, we neglect the small contribution to the background from atoms with $v_y(0)<\sqrt{4gw_0}$ which enter the laser beams only once and fall back out after being excited.)

The fountain apparatus is used to observe two-photon

transitions from the ground state $|b\rangle$, with Bohr frequency ω_b, to a metastable excited state $|a\rangle$, with Bohr frequency ω_a. We label the resonance frequency $\omega_{ab}=\omega_a-\omega_b$, and we assume that the atom decays from the upper level to the lower level at a rate $\gamma \leqq 10\ \mathrm{s}^{-1}$. Generalizing Eq. (A6) to the case of a moving atom, and using the effective transition matrix element (B10), we find the excitation probability

$$\rho_{aa}^{(2)}(t)=|D_{ab}|^2 I_1 I_2 \exp(-\gamma t)\left|\int_{-\infty}^{t} dt' f(t') \exp\left[-i\left[2\Omega t'-\Delta\mathbf{k}\cdot\mathbf{r}(t')+(\phi_1+\phi_2)-\int_{-\infty}^{t'} dt''\Delta\omega_{ab}(t'')+i\gamma t'/2\right]\right]\right|^2 . \quad (2.1)$$

Here D_{ab} is defined by Eq. (B11), the angular frequency $\Omega=\omega-\frac{1}{2}\omega_{ab}$ is the laser detuning from resonance, $\mathbf{r}(t)$ is the position of the atom at time t, and $f(t)$ is an envelope function given by

$$f(t)=\begin{cases}1, & 0\leqq t\leqq\tau \text{ and } T\leqq t\leqq T+\tau \\ 0 & \text{otherwise}.\end{cases} \quad (2.2)$$

The phase shift $\Delta\mathbf{k}\cdot\mathbf{r}(t)$ gives rise to a residual first-order Doppler effect. The frequency shift $\Delta\omega_{ab}(t)$ represents contributions from the ac Stark effect,[15] the second-order Doppler effect,[14] and the gravitational red shift.[16] The ac Stark shift occurs only when the atom interacts with the light field, and so it can be represented simply by $\Delta\omega_s f(t)$, where $\Delta\omega_s$ is given by Eq. (B12) and depends linearly on the laser intensities I_1 and I_2. In the laboratory reference frame, the second-order Doppler effect red shifts the atomic resonance frequency by an amount $-\omega_{ab}\mathbf{v}^2(t)/2c^2$. In addition, however, the gravitational red shift of the resonance frequency will be reduced by an amount $-\omega_{ab}\mathbf{g}\cdot\mathbf{r}(t)/c^2$ as the atom rises to apogee.[16,17] Hence, at both $t=0$ and $t=T+\tau$, ω_{ab} is red shifted compared to the resonance frequency of an atom that is simply at rest in the interaction region. Near the top of the atom's parabolic trajectory, however, where $\mathbf{v}(t)=0$, ω_{ab} is shifted towards the blue. We include all three effects by writing $\Delta\omega_{ab}(t)$ as

$$\Delta\omega_{ab}(t)=\Delta\omega_s f(t)-\frac{\omega_{ab}}{2c^2}[\mathbf{v}^2(t)+2\mathbf{g}\cdot\mathbf{r}(t)] . \quad (2.3)$$

To proceed, we evaluate Eq. (2.1) at $t=T+\tau$ and use Eq. (2.2) to break up the integral in Eq. (2.1) into two integrals corresponding to the two distinct interactions, and we perform the change of variable $t\rightarrow t+T$ in the latter. Furthermore, those parts of the total shift proportional to either $v_y(t)$ or $v_y^2(t)$ are significant only when $v_y(0)\gg\sqrt{4gw_0}$. When calculating the total shift *during* either of the two interactions, therefore, we may take $T\gg\tau$ and assume that the velocity of the atom is essentially constant. The final excitation probability will not depend on the phase factor $\exp[-i(\phi_1+\phi_2)]$, since it is common to both integrals. If we use the conservation of energy, $\mathbf{v}^2(t)-2\mathbf{g}\cdot\mathbf{r}(t)=\mathbf{v}^2(0)$, to rewrite the second term of Eq. (2.3), with all of these modifications Eq. (2.1) becomes

$$\begin{aligned}\rho_{aa}^{(2)}(T+\tau)=&|D_{ab}|^2 I_1 I_2 \exp[-\gamma(T+\tau)] \\ &\times\left|\int_0^{\tau} dt \exp\left[-i\left[2\Omega-\Delta\omega_s+\omega_{ab}\frac{\mathbf{v}^2(0)}{2c^2}-\Delta k_x v_x-\Delta k_y v_y(0)-\Delta k_z v_z+i\frac{\gamma}{2}\right]t\right]\right. \\ &+\exp[-i\Phi+i\Delta k_y v_y(0)\tau+\gamma T/2] \\ &\left.\times\int_0^{\tau} dt \exp\left[-i\left[2\Omega-\Delta\omega_s+\omega_{ab}\frac{\mathbf{v}^2(0)}{2c^2}-\Delta k_x v_x+\Delta k_y v_y(0)-\Delta k_z v_z+i\frac{\gamma}{2}\right]t\right]\right|^2 , \end{aligned} \quad (2.4)$$

where Δk_x, Δk_y, and Δk_z are the projections of $\Delta\mathbf{k}$ onto the x, y, and z axes, respectively, and

$$\Phi=2\Omega T-\Delta\omega_s\tau+\frac{\omega_{ab}}{2c^2}[v_x^2+v_z^2-\tfrac{1}{3}v_y^2(0)]T-(\Delta k_x v_x+\Delta k_z v_z)T . \quad (2.5)$$

The maximum interaction time is $\tau_{\max}=\sqrt{4w_0/g}$; if we assume that $\gamma\ll 2/\tau_{\max}$, then we can neglect damping during the interactions, and Eq. (2.4) becomes

$$\begin{aligned}\rho_{aa}^{(2)}(T+\tau)=&|D_{ab}|^2 I_1 I_2 \tau^2 \exp(-\gamma T)\left|j_0\left[\frac{1}{2}\left[2\Omega-\Delta\omega_s+\omega_{ab}\frac{\mathbf{v}^2(0)}{2c^2}-\Delta k_x v_x-\Delta k_y v_y(0)-\Delta k_z v_z\right]\tau\right]\right. \\ &\left.+\exp(-i\Phi+\gamma T/2) j_0\left[\frac{1}{2}\left[2\Omega-\Delta\omega_s+\omega_{ab}\frac{\mathbf{v}^2(0)}{2c^2}-\Delta k_x v_x+\Delta k_y v_y(0)-\Delta k_z v_z\right]\tau\right]\right|^2 , \end{aligned} \quad (2.6)$$

where $j_0(x)=\sin(x)/x$ is the zeroth-order spherical Bessel function.

Each term in Eq. (2.6) has a simple, intuitive explanation. The first Bessel function is proportional to the probability amplitude for excitation as the atom passes upwards through the (rectangular) light fields, while the second corresponds to that of the downward passage. The frequency corrections in the arguments of the Bessel functions are the ac Stark shift, the second-order Doppler red shift, and the residual first-order Doppler shift. The relative phase Φ is just the phase that the atom accrues as it travels between the interactions, and it is this which gives rise to the Ramsey fringes in the excitation probability. In Φ, note the characteristic reduction of the ac Stark shift by τ/T, since the two-photon resonance frequency is shifted only during the short interaction time τ.[2,14] The third term in Φ is the sum of the second-order Doppler shift and the gravitational red shift after integration over the atom's parabolic trajectory.[12] In the case where $v_x^2+v_z^2 \ll v_y^2(0)$, this results in a net *blue* shift of the Ramsey pattern.

We now consider in detail the consequences of a misalignment of the laser beams by the small relative angle $\Delta\theta=\sqrt{2\Delta k_z/k}$. The resulting two-photon recoil shift $(\hbar\omega_{ab}/Mc^2)(\Delta\theta/2)^2$, which we have not explicitly included in Eq. (2.6), remains below two parts in 10^{17} for $\Delta\theta<100$ μrad. The same is true for the residual first-order Doppler shift due to horizontal motion along the laser beams, $\Delta k_z v_z=\omega_{ab}(v_z/c)(\Delta\theta/2)^2$, and the corresponding second-order Doppler shift, $\omega_{ab}v_z^2/2c^2$, if $v_z<2$ m/s. However, in order to keep the residual first-order Doppler shift due to horizontal motion transverse to the beams, $\Delta k_x v_x=\omega_{ab}(v_x/c)(\Delta\theta/2)$, below two parts in 10^{17}, the apparatus would have to restrict $|v_x|$ to less than 1 mm/s even if the horizontal misalignment is only 10 μrad. In what follows we assume that these conditions have been met, and we ignore any first-order or second-order shifts caused by nonzero horizontal velocities. However, most of the atoms which will eventually contribute to the central interference signal are evenly distributed over the velocity range $0<v_y(0)<g/\gamma$, and so at this point we cannot ignore the $v_y(0)$ contributions to the second-order shifts in Eq. (2.6). But we can approximate $v_y(0)$ by the magnitude of the total velocity at $t=0$, which we denote simply as v from now on.

If $\Delta k_y v,v^2/2c^2 \ll 1/\tau_{\max}$, then the Doppler shifts of the broad background terms are small compared to their widths. Therefore, near line center, both of the Bessel functions in Eq. (2.6) may be replaced simply by 1, and we finally obtain

$$\rho_{aa}^{(2)}(T+\tau)=\rho_{aa}^{(2)}(\tau)[1+\exp(-\gamma T)+2\exp(-\gamma T/2)\cos(2\Omega T-\Delta\omega_s\tau-\omega_{ab}Tv^2/6c^2)]\ , \tag{2.7}$$

where $\tau\approx 2w_0/v$, $T\approx 2v/g$, and $\rho_{aa}^{(2)}(\tau)=|D_{ab}|^2 I_1 I_2 \tau^2$ is the transit-time-limited Doppler-free two-photon excitation probability at line center.

Two of the most interesting advantages of the atomic fountain technique become apparent upon close examination of Eq. (2.7). First of all, the phases ϕ_1 and ϕ_2 have completely disappeared from the final result, because the atoms interact twice with the same counterpropagating beams. In an ordinary Ramsey experiment,[2] an additional shift $-(\phi_1+\phi_2-\phi_3-\phi_4)$ would appear in Φ, with each ϕ coming from two laser beams in two separate interaction regions,[14] and great care must be taken to avoid cavity phase shifts of the central peak. Second, the atomic fountain completely avoids residual first-order Doppler shifts due to vertical motion through slightly misaligned beams because any net phase shift accumulated as the atom ascends is canceled by an equal and opposite shift acquired during the descent.

III. THE VELOCITY-AVERAGED EXCITATION PROBABILITY

In order to determine the signal line shape for the atomic fountain, we must average Eq. (2.7) over the atomic velocity distribution. We define Φ_b as the flux of ground-state atoms in $s^{-1}sr^{-1}$ emitted upward by the source located at $y=-h$ towards the interaction region at $y=0$. An atom with speed v at the nozzle must be emitted into an effective solid angle $\eta(v)$ in order to pass twice through the exciting laser fields and then reach the detector. We assume that for the atoms which contribute substantially to the central interference signal, $\mathbf{v}$ is essentially vertical, so that $\eta(v)$ is small and does not depend strongly on speed; we denote these quantities simply as v and η in the following discussion. We define the velocity distribution $n(v)dv$ as the number of atoms per unit distance with speeds in the range v to $v+dv$ emitted by the source into the solid angle η. Above the source the atoms are no longer in thermal equilibrium, and we may treat the motion of each particle as that of a simple projectile subject to the force of gravity. Those atoms with $v<\sqrt{2g(y+h)}$ at the source are too slow to rise to vertical position y. The velocity distribution at arbitrary y, $n'(v,y)dv$, may be deduced from $n(v)dv$ simply by making the change of variable $v^2\rightarrow v^2+2g(y+h)$:

$$n(v)dv\rightarrow\frac{n(\sqrt{v^2+2g(y+h)})}{\sqrt{v^2+2g(y+h)}}vdv\equiv n'(v,y)dv\ . \tag{3.1}$$

The atomic current distribution at arbitrary y, $j_b(v,y)dv$, is proportional to $n'(v,y)vdv$[18] and is normalized so that the integral of $j_b(v,-h)dv$ over all positive speeds yields $\Phi_b\eta$. Therefore, if we assume that $n(v)$ corresponds to a Maxwellian velocity distribution,[18] then the atomic current distribution in the interaction region is

$$j_b(v,0)dv=\Phi_b\eta(2v^2/v_0^4)\sqrt{v^2+2gh}\times\exp(-v^2/v_0^2-2gh/v_0^2)dv\ , \tag{3.2}$$

where v_0 is the most probable atomic speed at $y=-h$.[18] [We have corrected an obvious sign error in Eq. (2) of Ref. 3.] Now, if we multiply Eq. (2.7) by Eq. (3.2) and in-

tegrate over velocity, we obtain for the signal line shape

$$\Delta J_a(\Omega)=2\Phi_b\eta\rho_{aa}^{(2)}(\tau_0)K(\Omega)\ , \tag{3.3}$$

where the line-shape function $K(\Omega)$ is given by

$$K(\Omega)=2\exp(-2gh/v_0^2)\times\int_{u_{\min}}^{\infty}du\sqrt{u^2+2gh/v_0^2}\exp(-\gamma T_0u/2-u^2)\times\cos\left[2\Omega T_0u-\Delta\omega_s\frac{\tau_0}{u}-\omega_{ab}T_0\frac{v_0^2}{6c^2}u^3\right]. \tag{3.4}$$

Here $\tau_0=2w_0/v_0$, $T_0=2v_0/g$, and $u_{\min}=\sqrt{16gw_0/v_0^2}$. The lower limit of integration has been chosen so as to ignore the broad background introduced by very slow atoms which rise less than three beam diameters above the laser field. In a more precise treatment these atoms would require special consideration: to first order in $u_{\min}$, they simply add a small constant background $2\sqrt{2gh/v_0^2}\exp(-2gh/v_0^2)u_{\min}$ to $K(\Omega)$ near line center. We have scaled $K(\Omega)$ so that in a standard Ramsey geometry, when damping between the interactions can be ignored,, $K(0)=1$. In the limit where $u_{\min}\ll 1$ and $\gamma T_0/2\gg 1$, we neglect all shifts and expand the Maxwellian factor $\sqrt{u^2+2gh/v_0^2}\exp(-u^2)$ to second order in u. Then the central interference signal becomes

$$\Delta J_a(\Omega)=4\Phi_b\eta\rho_{aa}^{(2)}(\tau_0)\frac{2}{\gamma T_0}\left[\frac{2gh}{v_0^2}\right]^{1/2}\times\exp(-2gh/v_0^2)\frac{(\gamma/4)^2}{\Omega^2+(\gamma/4)^2}\times\left[1+\frac{2(1-v_0^2/4gh)}{(\gamma T_0/2)^2}\times(\gamma/4)^2\frac{3\Omega^2-(\gamma/4)^2}{[\Omega^2+(\gamma/4)^2]^2}\right]. \tag{3.5}$$

The leading term in this approximation is just a Lorentzian. The correction factor due to the second term in the large parentheses is $-2(1-v_0^2/4gh)/(\gamma T_0/2)^2$ at line center; hence, for a truly Maxwellian velocity distribution at the source, a fountain with an interaction region placed at a height $h=v_0^2/4g$ above the nozzle will provide an almost perfectly Lorentzian line shape. The lowest-order Lorentzian term in Eq. (3.5) does not depend on the exact shape of the velocity distribution, as can be shown by expanding $n'(v,0)$ in a Taylor series about $v=0$, noting that $n'(0,0)=0$, and then keeping the first-order term. At line center, the ratio of the Doppler-free two-photon background to this signal is approximately $\frac{1}{4}+(1+v_0^2/8gh)\sqrt{\pi}\gamma T_0/8$.

IV. EXAMPLE: THE HYDROGEN 1S-2S TRANSITION

To consider a tractable example of particular interest, we apply the results of our analysis to the $1S$-$2S$ transition of atomic hydrogen.[4] The two-photon decay rate of the metastable $2S$ state[19] is $\gamma=8.23\ \mathrm{s}^{-1}$, so that the natural linewidth of the $1S$-$2S$ transition is 0.655 Hz (at 243 nm). We assume that the laser fields have parallel linear polarizations, and we choose the atomic axis of quantization along the direction of the electric field. In this case, the two-photon transition amplitude defined by Eq. (B13) has been computed to be[20]

$$M_{ab}^{12}=11.78\ , \tag{4.1}$$

giving for (B11)

$$D_{ab}=4.63\ \mathrm{cm^2\,J^{-1}}\ . \tag{4.2}$$

If the two laser beams have the same intensity I, then the ac Stark effect (B12) can be obtained using[21]

$$(M_{aa}^{11}+M_{aa}^{22})-(M_{bb}^{11}+M_{bb}^{22})=53.35\ , \tag{4.3}$$

resulting in a shift of the $1S$-$2S$ transition frequency (at 243 nm) by

$$\frac{\Delta\omega_s}{4\pi}=1.67I\ \mathrm{Hz\,W^{-1}\,cm^2}\ . \tag{4.4}$$

We assume that the hydrogen atoms at the source have been cooled to a temperature of 1 mK. In principle, radiation pressure cooling of hydrogen atoms could be accomplished very effectively with laser light at the 121.6-nm $L\alpha$ line. Unfortunately, there are still no suitable tunable vacuum ultraviolet lasers available, and so far nonlinear four-wave mixing has produced only short pulses of modest power.[22] On the other hand, once the atoms are precooled to liquid helium temperature by conventional means,[23] a hydrogen atom can be stopped over a very short distance by resonant scattering of only about 100 $L\alpha$ photons. Alternatively, the atoms could be cooled into the millikelvin range with the help of a dilution refrigerator, or inhomogeneous magnetic fields might be employed for velocity reduction and selection.

We assume that the standing-wave light field is located at a height $h=v_0^2/2g=85$ cm above the source and has a vertical thickness $2w_0=100\ \mu$m. It may be desirable to expand the beams in the horizontal direction to a width of a few millimeters. Since the mass of the hydrogen atom is so small, if we restrict the horizontal speed v_x to less than 1 mm/s, then according to the Heisenberg uncertainty principle we cannot localize the x component of the atom's position to better than 60 μm.

The average resonant excitation probability for an atom passing twice through this interaction region is approximately $10^{-8}I^2\ \mathrm{cm^4\,W^{-2}}$; the corresponding count rate is obtained by multiplying this number by $\Phi_b\eta$. The continuous curve in Fig. 2 shows the signal line shape $K(\Omega)$ as predicted by a full numerical integration of Eq. (3.4) for $I=1\ \mathrm{W/cm^2}$, including a small constant background $2\sqrt{2gh/v_0^2}\exp(-2gh/v_0^2)u_{\min}$. There is a slight net blue shift of 0.02 Hz mainly due to the second-order Doppler effect and the gravitational red shift. The ac Stark shift remains below 0.02 Hz up to intensities of 30 $\mathrm{W/cm^2}$, although the line shape becomes more asymmetric at higher intensities. The limiting assumptions made in the derivation of Eq. (3.5) are not well satisfied here, since the hydrogen atom is so light. Nevertheless, the Lorentzian

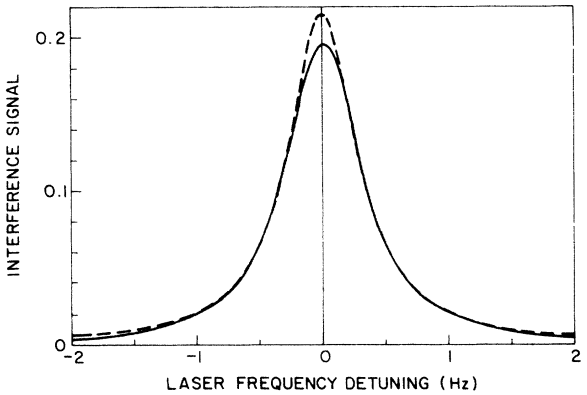

FIG. 2. Interference signal expected for the hydrogen $1S$-$2S$ transition (source temperature 1 mK, $h=85$ cm, $2w_0=100$ μm). The continuous curve gives the result obtained by numerical integration of Eq. (3.4). The dashed curve shows the Lorentzian approximation predicted by the leading-order term in Eq. (3.5).

line-shape approximation, given by the leading-order term in Eq. (3.5) and illustrated by the dashed curve in Fig. 2, describes the line shape fairly well. The 8% difference in signal height between the exact and Lorentzian line profiles is due almost entirely to our neglect of the correction term in Eq. (3.5). In the chosen units, the Doppler-free background reaches about 0.4 at its center.

Of course, it will be important to shield the hydrogen atoms from perturbing electric fields so as to avoid dc Stark shifts. For a small electric field E, Stark mixing of the $2S_{1/2}$ state with the closely adjacent $2P_{1/2}$ and $2P_{3/2}$ states[24] shifts the $2S$ level upward by about $3600E^2$ Hz V^{-2} cm^2. In order to keep the resulting blue shift of the $1S$-$2S$ transition below 0.02 Hz (at 243 nm), any external electric fields would need to be reduced below 3.3 mV/cm.

Magnetic fields pose a less serious problem since both the $1S$ and the $2S$ states have the same magnetic moment at low fields, and the selection rules $\Delta F=0$ and $\Delta m_F=0$ permit transitions only between corresponding Zeeman sublevels.[25] Since the $1S$ and $2S$ levels have different hyperfine coupling strengths, the $m_F=0$ transitions are field sensitive, and magnetic hyperfine decoupling gives a shift of about 10^4B^2 Hz G^{-2}. Hence, any external magnetic fields must be smaller than 2 mG if these shifts are to remain below 0.02 Hz (at 243 nm). But the line components with $m_F=\pm1$ will exhibit *zero* Zeeman shifts up to much larger fields. Motional Stark shifts due to the electric field observed by an atom moving through a dc magnetic field are also negligible up to about 1 kG as long as the atoms are cooled to millikelvin temperatures so that $v/c \leqq 10^{-8}$.

Thus, while formidable technical difficulties remain, the proposed fountain experiment promises a viable path towards the tantalizing goal of measuring the hydrogen $1S$-$2S$ transition with a natural-linewidth-limited resolution of better than one part in 10^{15}.

V. CONCLUSIONS

In conclusion, we have presented a semiclassical model exploring the merits of two-photon optical Ramsey spectroscopy of neutral atoms falling freely in a fountain. The proposed technique can provide simple interference signals of ultrahigh resolution without perturbing the atoms with trapping fields. We have investigated a number of systematic corrections, including the effects of beam misalignment, first-order and second-order Doppler shifts, gravitational red shifts, and ac Stark shifts, and we point out important advantages of the fountain geometry compared to more conventional excitation schemes. Continuing rapid progress in laser frequency stabilization techniques and laser cooling technology should make it possible to perform such an experiment in the near future.

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APPENDIX A: THE TWO-LEVEL OPTICAL BLOCH EQUATIONS

Consider an unperturbed atom described by an effective two-level Hamiltonian whose eigenvalues and eigenfunctions are known. In the basis in which the Hamiltonian is diagonal, let $|a\rangle$ and $|b\rangle$ denote the eigenstate vectors describing the upper and lower levels, respectively, and let $\hbar\omega_a$ and $\hbar\omega_b$ denote the corresponding energy eigenvalues. We allow the atom to interact with a c-number electromagnetic field through the interaction Hamiltonian $H_1(t)$, which not only causes transitions between the two states, but also shifts the Bohr frequencies ω_a and ω_b by the amounts $\Delta\omega_a(t)$ and $\Delta\omega_b(t)$. Using time-dependent perturbation theory in the rest frame of the atom we write the equations of motion for the Schrödinger picture probability amplitudes as[25,26]

$$\dot{c}_a(t)=-i[\omega_a+\Delta\omega_a(t)]c_a(t)-\frac{i}{\hbar}V_{ab}(t)c_b(t)\ , \tag{A1}$$

$$\dot{c}_b(t)=-i[\omega_b+\Delta\omega_b(t)]c_b(t)-\frac{i}{\hbar}V_{ba}(t)c_a(t)\ .$$

Here $c_a(t)$ is the probability amplitude for finding the atom in the unperturbed upper state at time t, $c_b(t)$ is the corresponding probability amplitude for the lower state, and $V_{ab}(t)=V_{ba}^*(t)$. For N-photon resonant transitions, $V_{ab}(t)$ is of order N in $H_1(t)$, but $\Delta\omega_a(t)$ and $\Delta\omega_b(t)$ are always quadratic in $H_1(t)$.

The method of obtaining the appropriate two-level optical Bloch equations from the above equations of motion is well known.[26] From Eqs. (A1) we identify the effective two-level Hamiltonian $H(t)=H_0+V(t)$, where

$$H_0=\begin{bmatrix}\hbar\omega_a & 0\\ 0 & \hbar\omega_b\end{bmatrix}, \tag{A2}$$

$$V(t)=\begin{bmatrix}\hbar\Delta\omega_a(t) & V_{ab}(t)\\ V_{ba}(t) & \hbar\Delta\omega_b(t)\end{bmatrix}. \tag{A3}$$

The density operator ρ has the matrix elements $\rho_{nm}(t)=\langle n\,|\,\rho(t)\,|\,m\rangle=c_n(t)c_m^*(t)$, where $n,m=a,b$, and in the atomic rest frame it obeys the equation of motion

$$i\hbar\dot{\rho}=[H,\rho]+i\hbar\Gamma(\rho)\ . \tag{A4}$$

Here the matrix $\Gamma(\rho)$ describes relaxation due to Wigner-Weisskopf decay and dephasing processes such as collisions. We are particularly interested in the case where $|b\rangle$ is the ground state, and $|a\rangle$ is a metastable state which may decay to $|b\rangle$ after a mean lifetime γ^{-1}. If we neglect collisions, we have

$$\Gamma(\rho)=-\begin{bmatrix}\gamma\rho_{aa} & \frac{\gamma}{2}\rho_{ab}\\ \frac{\gamma}{2}\rho_{ba} & -\gamma\rho_{aa}\end{bmatrix}. \tag{A5}$$

We define the resonance frequency $\omega_{ab}=\omega_a-\omega_b$ and the total shift $\Delta\omega_{ab}(t)=\Delta\omega_a(t)-\Delta\omega_b(t)$. If the initial conditions are $\rho_{aa}(t)\to 0$ and $\rho_{bb}(t)\to 1$ as $t\to-\infty$, then solving Eq. (A4) for $\rho_{aa}(t)$ and $\rho_{bb}(t)$ to second order in $V_{ab}(t)$ gives

$$\rho_{aa}^{(2)}(t)=\exp(-\gamma t)\left|\int_{-\infty}^{t}dt'\frac{V_{ab}(t')}{\hbar}\exp\left[i\omega_{ab}t'+i\int_{-\infty}^{t'}dt''\Delta\omega_{ab}(t'')+\gamma t'/2\right]\right|^2,$$

$$\rho_{bb}^{(2)}(t)=1-\rho_{aa}^{(2)}(t)\ . \tag{A6}$$

APPENDIX B: AN EFFECTIVE TWO-LEVEL HAMILTONIAN FOR SEMICLASSICAL TWO-PHOTON TRANSITIONS

Here we employ conventional techniques to derive an effective semiclassical Hamiltonian for resonant two-photon transitions.[1,27] Unlike a quantum electrodynamic treatment describing each of the counterpropagating laser beams with photon number states, our model accounts for spatial interference effects in the standing-wave electric field. We discuss below the conditions under which this interference can be neglected, and our result reduces to the form usually found in the literature.

In the EDA, we consider resonant semiclassical two-photon transitions between the two states $|a\rangle$ and $|b\rangle$ of the atom illustrated in Fig. 3. The electric dipole matrix element between states $|a\rangle$ and $|b\rangle$ is zero, but non-resonant one-photon electric dipole transitions from $|a\rangle$ and $|b\rangle$ to intermediate states $|n\rangle$ are allowed. For convenience we first determine the equations of motion for the interaction picture probability amplitudes, given in terms of those in the Schrödinger picture by $C_k(t)=c_k(t)\exp(i\omega_k t)$. Time-dependent perturbation theory yields[25,26]

$$\dot{C}_k(t)=\frac{1}{i\hbar}\sum_n\langle k\,|\,H_1(t)\,|\,n\rangle\exp(i\omega_{kn}t)C_n(t)\ , \tag{B1}$$

where $\omega_{kn}=\omega_k-\omega_n$. The sum over n includes an integration over continuum states. In the EDA the interaction Hamiltonian is given by[26]

$$H_1(t)=-ea_0\mathbf{d}\cdot\mathbf{E}(\mathbf{r},t)\ , \tag{B2}$$

where $e<0$ is the charge of the electron, a_0 is the Bohr radius, $ea_0\mathbf{d}$ is the electric dipole operator,[28] and $\mathbf{E}(\mathbf{r},t)$ is the total classical electric field at $\mathbf{r}$, the position of the atomic nucleus. In the case where the exciting laser bandwidths are much smaller than the resolution of the experiment (which may be limited by either the natural linewidth of the transition or the finite interaction time), the electric field may be written as the sum of two monochromatic fields with angular frequencies ω_1 and ω_2 tuned so that $\omega_1+\omega_2\approx\omega_{ab}$ and complex vector amplitude functions $\mathbf{E}_1(\mathbf{r},t)$ and $\mathbf{E}_2(\mathbf{r},t)$:

$$\mathbf{E}(\mathbf{r},t)=\tfrac{1}{2}\mathbf{E}_1(\mathbf{r},t)\exp(-i\omega_1 t)+\text{c.c.}$$
$$+\tfrac{1}{2}\mathbf{E}_2(\mathbf{r},t)\exp(-i\omega_2 t)+\text{c.c.} \tag{B3}$$

The equations of motion for the probability amplitudes $C_a(t)$ and $C_b(t)$ are therefore

$$\dot{C}_a(t)=i\frac{ea_0}{2\hbar}\sum_n\mathbf{d}_{an}\cdot\mathbf{E}_1(\mathbf{r},t)\exp[i(\omega_{an}-\omega_1)t]C_n(t)$$
$$+i\frac{ea_0}{2\hbar}\sum_n\mathbf{d}_{an}\cdot\mathbf{E}_1^*(\mathbf{r},t)\exp[i(\omega_{an}+\omega_1)t]C_n(t)$$
$$+(1\to 2)\ , \tag{B4}$$

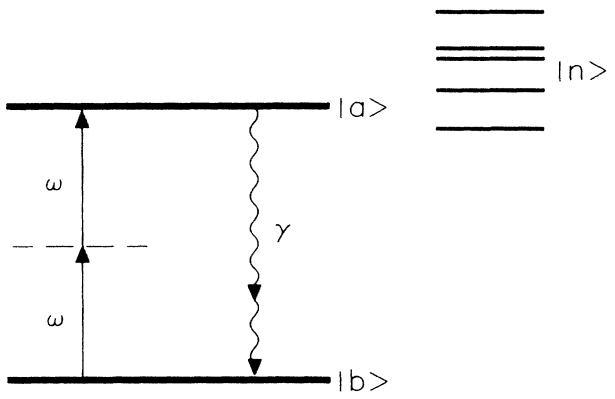

FIG. 3. Atomic energy level diagram for resonant two-photon transitions.

where $\mathbf{d}_{an}=\langle a \mid \mathbf{d} \mid n \rangle$. The expression for $C_b(t)$ is identical to Eq. (B4), except that $a \rightarrow b$. We cannot yet make the RWA,[26] since the frequencies ω_n are all far off resonance. (In the RWA, we neglect all but slowly varying terms, i.e., we ignore all terms with a very rapidly oscillating time dependence.) However, we anticipate that $C_a(t)$ and $C_b(t)$ will be slowly varying, so that the only terms in the equation of motion for the probability amplitude $C_n(t)$ of the intermediate state $\mid n \rangle$ which will eventually contribute to a two-photon resonance are those proportional to $C_a(t)$ or $C_b(t)$. Then

$$\dot{C}_n(t) \approx i\frac{ea_0}{2\hbar}\mathbf{d}_{na}\cdot\mathbf{E}_1(\mathbf{r},t)\exp[-i(\omega_{an}+\omega_1)t]C_a(t)+i\frac{ea_0}{2\hbar}\mathbf{d}_{na}\cdot\mathbf{E}_1^*(\mathbf{r},t)\exp[-i(\omega_{an}-\omega_1)t]C_a(t)$$

$$+(\text{terms with } 1\rightarrow 2)+(\text{similar terms with } a\rightarrow b)\ . \tag{B5}$$

If the lasers are tuned very near to the two-photon resonance, then $C_a(t)$ and $C_b(t)$ oscillate slowly compared to frequencies such as $\omega_{an}\pm\omega_1$. Hence, if we can make the adiabatic approximation[15] $\mid \dot{\mathbf{E}}_j(\mathbf{r},t) \mid \ll \mid \omega \mathbf{E}_j(\mathbf{r},t) \mid$, where $\omega=\omega_{mn}\pm\omega_j$, $j=1$ or 2 and $m=a$ or b [i.e., if we can assume that neither $\mathbf{E}_1(\mathbf{r},t)$ nor $\mathbf{E}_2(\mathbf{r},t)$ has any significant high-frequency components], and if $\mid \mathbf{E}_1(\mathbf{r},t) \mid, \mid \mathbf{E}_2(\mathbf{r},t) \mid \rightarrow 0$ as $t\rightarrow-\infty$, then we can integrate Eq. (B5) directly. After we substitute the result into Eq. (B4), make the RWA, and convert the interaction picture probability amplitudes into those of the Schrödinger picture, we obtain Eqs. (A1) with

$$\begin{aligned}\Delta\omega_a(t)&=\Delta\omega_a^{11}(t)+\Delta\omega_a^{22}(t)+\Delta\omega_a^{12}(t)\exp[-i(\omega_1-\omega_2)t]+\Delta\omega_a^{21}(t)\exp[+i(\omega_1-\omega_2)t]\ ,\\ V_{ab}(t)&=V_{ab}^{11}(t)\exp(-i2\omega_1 t)+V_{ab}^{22}(t)\exp(-i2\omega_2 t)+[V_{ab}^{12}(t)+V_{ab}^{21}(t)]\exp[-i(\omega_1+\omega_2)t]\ ,\end{aligned} \tag{B6}$$

where

$$\begin{aligned}\Delta\omega_a^{ij}(t)&=\left[\frac{ea_0}{2\hbar}\right]^2\sum_n\left[\frac{\mathbf{d}_{an}\cdot\mathbf{E}_j^*(\mathbf{r},t)\mathbf{d}_{na}\cdot\mathbf{E}_i(\mathbf{r},t)}{\omega_{an}+\omega_i}+\frac{\mathbf{d}_{an}\cdot\mathbf{E}_i(\mathbf{r},t)\mathbf{d}_{na}\cdot\mathbf{E}_j^*(\mathbf{r},t)}{\omega_{an}-\omega_j}\right]\ ,\\ \frac{V_{ab}^{ij}(t)}{\hbar}&=\left[\frac{ea_0}{2\hbar}\right]^2\sum_n\frac{\mathbf{d}_{an}\cdot\mathbf{E}_j(\mathbf{r},t)\mathbf{d}_{nb}\cdot\mathbf{E}_i(\mathbf{r},t)}{\omega_{bn}+\omega_i}\ .\end{aligned} \tag{B7}$$

The expression for $\Delta\omega_b(t)$ may be obtained from the first of Eqs. (B6) and (B7) by making the substitution $a\rightarrow b$. Physically, the two-photon transition potential $V_{ab}(t)$ and the small ac Stark shifts $\Delta\omega_a(t)$ and $\Delta\omega_b(t)$ all arise from two concatenated first-order virtual electric dipole transitions: the first is from the initial level to a nonresonant intermediate level $\mid n \rangle$ and the second is either to the other state (stimulated two-photon absorption or emission) or back to the same state (ac Stark shift).

When $\omega_1-\omega_2$ is much larger than the resolution of the experiment, the last two terms in the first of Eqs. (B6) are rapidly varying and can be neglected. In this work, however, we are primarily concerned with the important case where $\omega_1=\omega_2=\omega$; after this simplification Eqs. (B6) become

$$\begin{aligned}\Delta\omega_a(t)&=\left[\frac{ea_0}{2\hbar}\right]^2\sum_n\left[\frac{\mid \mathbf{d}_{an}\cdot[\mathbf{E}_1^*(\mathbf{r},t)+\mathbf{E}_2^*(\mathbf{r},t)] \mid^2}{\omega_{an}+\omega}+\frac{\mid \mathbf{d}_{an}\cdot[\mathbf{E}_1(\mathbf{r},t)+\mathbf{E}_2(\mathbf{r},t)] \mid^2}{\omega_{an}-\omega}\right]\ ,\\ \frac{V_{ab}(t)}{\hbar}&=\left[\frac{ea_0}{2\hbar}\right]^2\exp(-i2\omega t)\sum_n\frac{\mathbf{d}_{an}\cdot[\mathbf{E}_1(\mathbf{r},t)+\mathbf{E}_2(\mathbf{r},t)]\mathbf{d}_{nb}\cdot[\mathbf{E}_1(\mathbf{r},t)+\mathbf{E}_2(\mathbf{r},t)]}{\omega_{bn}+\omega}\end{aligned} \tag{B8}$$

with a similar expression for $\Delta\omega_b(t)$.

Here we have treated the laser light as a purely classical electromagnetic field, and we have used the term "two-photon transition" to denote a resonant second-order semiclassical field-atom interaction in the electric dipole approximation. Since electromagnetic field quantization is important only in phenomena where vacuum field-atom interactions play a significant role,[29,30] we expect a full quantum electrodynamic analysis of the same problem to yield Eqs. (B8). This will not happen, however, if the quantum laser field is described in the "fuzzy-ball" photon model by simple counterpropagating photon number states. In the case of two-photon absorption from counterpropagating beams which have the same frequency, this model predicts a degenerate final state: the possibility exists to absorb two photons from the first beam, or two photons from the second beam, or one photon from each beam (through two different paths). The probability amplitudes for these three processes are usually calculated separately, squared, and then added together to obtain the total absorption probability.[15,31–33] This procedure results in an expression which does not depend on spatial properties of the laser field, and therefore it neglects any interference between the counterpropagating beams.

Under certain circumstances, however, this can be a serious omission; for example, an atom traveling along a node of a standing-wave electric field will *not* be excited and will *not* suffer a level shift. Clearly, the fuzzy-ball photon model fails here,[29] because the state vectors for the field have completely unknown relative phases and *cannot* interfere. If the electromagnetic field is quantized with the spatial boundary conditions satisfied by the corresponding classical field, however, as is done in Ref. 26 for the case of standing waves in a laser cavity, then the correct phases are automatically acquired and Eqs. (B8) are obtained, with the classical field amplitudes replaced by the appropriate electric field operators and the purely atomic state vectors replaced by field-atom state vectors.

Nevertheless, in most situations of practical interest the interference between the counterpropagating beams can be neglected,[34] and under these circumstances we recover the much simpler formula obtained using the fuzzy-ball photon model described above. Consider an atom with velocity $\mathbf{v}=v_y\hat{\mathbf{y}}+v_z\hat{\mathbf{z}}$ interacting with a standing-wave light field of thickness $2w_0$ consisting of two counterpropagating laser beams with propagation vectors $\mathbf{k}_1=k\hat{\mathbf{z}}$ and $\mathbf{k}_2=-k\hat{\mathbf{z}}$, where $kc=2\pi c/\lambda=\omega$ is the frequency of each laser. The number of standing-wave nodes that the atom crosses during the interaction is given by $2v_z\tau/\lambda$, where $\tau=2w_0/v_y$ is the interaction time. If $v_z/v_y \gg \lambda/4w_0$, then the atom traverses many nodes, and as long as the vast majority of atoms have velocities which satisfy this inequality, an average of the excitation probability given by Eq. (A6) over the atomic velocity distribution will eliminate the interference between the counterpropagating beams.

Even if velocities in the z direction are so tightly restricted that $v_z/v_y \leqq \lambda/4w_0$, the interference effects will average out for slow atoms because of the Heisenberg uncertainty principle. Our ability to localize the z component of the atom's position is limited by the inequality $\Delta z \geqq \hbar/Mv_z$, so that $\Delta z \geqq (2\hbar/Mv_y)(2w_0/\lambda)$. Hence, if $\Delta z \gg \lambda$, each atom will sample a large region of the standing-wave light field.

A further consequence of the averaging process over v_z is that the two Doppler-broadened terms due to second-order interactions with only one of the two beams will be very small at line center compared to the Doppler-free term caused by the interaction with both beams.[31,32] It is this latter term, arising from $V_{ab}^{12}(t)$ and $V_{ab}^{21}(t)$ in the second of Eqs. (B6), that we are concerned with in the fountain experiment, and so hereafter we neglect the contributions from the Doppler-broadened terms.

In writing our results below, we have assumed a particular form for the complex electric field amplitude $\mathbf{E}_j(\mathbf{r},t)$:

$$\mathbf{E}_j(\mathbf{r},t)=\boldsymbol{\epsilon}_j E_j(t)U_j(\mathbf{r})\exp[i(\mathbf{k}_j\cdot\mathbf{r}-\phi_j)] , \tag{B9}$$

where $\boldsymbol{\epsilon}_j$ is a complex polarization vector having unit magnitude, $E_j(t)$ is a real, slowly varying envelope function having the dimensions of an electric field, $U_j(\mathbf{r})$ is a dimensionless complex function describing the geometry of the light field, $\mathbf{k}_j$ is the propagation vector, and ϕ_j is a phase. Finally, then, for two-photon absorption from counterpropagating laser beams having the same frequency, the effective stimulated absorption-emission matrix element $V_{ab}(t)$ becomes

$$\frac{V_{ab}(t)}{\hbar}=D_{ab}\tfrac{1}{2}\epsilon_0 cE_1(t)E_2(t)U_1(\mathbf{r})U_2(\mathbf{r}) \times\exp i[(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{r}-2\omega t-(\phi_1+\phi_2)] , \tag{B10}$$

where

$$D_{ab}=\frac{1}{\pi^2\hbar c}\left[\frac{\alpha}{2R_\infty}\right]^3\frac{1}{3}M_{ab}^{12} \tag{B11}$$

and R_∞ is the Rydberg constant. For the ac Stark shift of the resonance frequency ω_{ab} we obtain

$$\Delta\omega_s(t)=\frac{1}{\pi^2\hbar c}\left[\frac{\alpha}{2R_\infty}\right]^3 \times\tfrac{1}{3}[(M_{aa}^{11}-M_{bb}^{11})\tfrac{1}{2}\epsilon_0 cE_1^2(t)\,|\,U_1(\mathbf{r})\,|^2 +(M_{aa}^{22}-M_{bb}^{22})\tfrac{1}{2}\epsilon_0 cE_2^2(t)\,|\,U_2(\mathbf{r})\,|^2] . \tag{B12}$$

The dimensionless two-photon transition amplitudes in Eqs. (B11) and (B12) are defined as

$$M_{ab}^{12}=3\pi cR_\infty\sum_n\frac{\boldsymbol{\epsilon}_1\cdot\mathbf{d}_{an}\boldsymbol{\epsilon}_2\cdot\mathbf{d}_{nb}+\boldsymbol{\epsilon}_2\cdot\mathbf{d}_{an}\boldsymbol{\epsilon}_1\cdot\mathbf{d}_{nb}}{\omega_{bn}+\omega} , \tag{B13}$$

$$M_{aa}^{jj}=3\pi cR_\infty\sum_n\left[\frac{|\,\boldsymbol{\epsilon}_j\cdot\mathbf{d}_{an}\,|^2}{\omega_{an}-\omega}+\frac{|\,\boldsymbol{\epsilon}_j^*\cdot\mathbf{d}_{an}\,|^2}{\omega_{an}+\omega}\right] . \tag{B14}$$

The expression for M_{bb}^{jj} is the same as Eq. (B14) with $a\rightarrow b$. The field-dependent factors in Eq. (B12), $\frac{1}{2}\epsilon_0 cE_j^2(t)\,|\,U_j(\mathbf{r})\,|^2$ $(j=1,2)$, are simply the intensities $I_j(\mathbf{r},t)$ of the two laser beams.

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