Two-photon Doppler-free spectroscopy of trapped atoms

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We describe the systematics of two-photon Doppler-free excitation of atoms confined in a cylindrical harmonic trap, at densities far from the regime of quantum degeneracy. The line shape is explained in terms of momentum transfer between the light beam and the trapped atoms. This formalism extends the semiclassical analysis of time-of-flight broadening. The results, which are general, were motivated by the development of two-photon Doppler-free spectroscopy of trapped atomic hydrogen. [S1050-2947(99)04406-6]

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I. INTRODUCTION

The intimate connection between the internal and external (translational) states of a radiating atom was first demonstrated by Einstein, who showed that the essential mechanism for maintaining thermal equilibrium between a thermal radiation field and the kinetic degrees of freedom of an atomic gas is the Doppler shift due to photon recoil [1]. The connection manifests itself in many other contexts, for instance in Mossbauer's analysis of recoil-free gamma ray-emission [2], and Dicke's proposal for suppressing Doppler broadening by confining the radiator to a distance short compared to its wavelength [3].

With the advent of techniques for laser cooling and trapping atoms, the effect of translational motion on the radiation process has taken on a new significance. Many of these phenomena were first studied for the case of trapped ions [4], and the analysis can often be extended to trapped atoms, with suitable allowance for the effects of the relatively soft confining potential of atom traps. Photoexcitation in the case of two-photon absorption, particularly in the Doppler-free configuration, involves rather different types of behavior. We have analyzed this behavior, motivated by the development of two-photon spectroscopy of trapped atomic hydrogen [5,6]. The transition observed, $1S \rightarrow 2S$, plays a prominent role in hydrogen spectroscopy [7], in precision tests of basic theory [8], and as a probe for studying Bose-Einstein condensation in spin-polarized hydrogen [9]. Due to the long lifetime of the metastable $2S$ state, the natural linewidth for this transition is only 1 Hz. By employing trapped atoms, it appears that this resolution may be experimentally feasible. Consequently, a detailed understanding of the radiation process is essential.

We present here an analysis of two-photon excitation of a trapped atomic gas that is far from quantum degeneracy. The discussion is confined to the so-called Doppler-free configuration in which two photons are absorbed from a standing wave oscillating at half the transition frequency [10,11]. Other than this, the discussion is general.

The system is described in Sec. II. In Sec. III the transition amplitude for two-photon absorption is calculated, and in Sec. IV the physical picture of momentum transfer from the laser beam to the atom is developed. The results are applied to spectroscopy with a weakly diverging laser beam (the limit of long Rayleight length) in Sec. V, and the photoexcitation line shapes under various conditions are described in Sec. VI. Section VII presents some concluding discussion.

II. THE SYSTEM

We consider Doppler-free two-photon excitation of atoms confined by a trapping potential, taking into account the quantum behavior of the translational motion. The trap has cylindrical symmetry, and is coaxial with a radiation field in the fundamental Gaussian mode.

The system is described by the Hamiltonian

$$H = H_o + H_{\rm cm} + V, \tag{1}$$

where H_o and $H_{\rm cm}$, respectively, describe the internal and translational states of the atom, and V describes the interaction of atom and radiation field. The eigenstates of H_o are taken to be the ground state, $|A\rangle$, a set of intermediate states with opposite parity, $|B\rangle$, and the final state, $|C\rangle$. States $|A\rangle$ and $|C\rangle$ are coupled to $|B\rangle$ by dipole matrix elements μ_{ab} and μ_{bc}, respectively. The electronic transition frequencies of H_o are in the form $\omega_{ba} \equiv (E_b - E_a)/\hbar$.

It is convenient to describe the center-of-mass motion using Cartesian coordinates with translational states $|n_x, n_y, n_z\rangle$, where n_x, n_y, and n_z are quantum numbers. For the purpose of discussion we assume that the trap is harmonic and cylindrically symmetric: extension of the argument to other geometries is straightforward. The Hamiltonian for the atom's center of mass motion is

$$H_{\rm cm} = \sum_{j=x,y,z} \left(\frac{P_j^2}{2m} + \frac{1}{2} m \Omega_j^2 R_j^2 \right) = \sum_{j=x,y,z} \hbar \Omega_j (n_j + 1/2), \tag{2}$$

where $\Omega_x = \Omega_y \equiv \Omega$ is the radial angular frequency. It is useful to introduce the parameter α:

$$\alpha \equiv \sqrt{m\Omega/\hbar}. \tag{3}$$

The rms momentum is $\langle p_j^2\rangle^{1/2}=\hbar\alpha\sqrt{j+1/2}$, and the rms size of the wave functions is $\langle x_j^2\rangle^{1/2}=\sqrt{j+1/2}/\alpha$.

In the dipole approximation, the atom-radiation interaction is

$$V(\mathbf{r},t)=-\boldsymbol{\mu}\cdot\mathbf{E}(\mathbf{r},t), \tag{4}$$

where $\boldsymbol{\mu}$ is the dipole operator and $\mathbf{E}(\mathbf{r},t)$ is the real electric field amplitude which is taken to oscillate at frequency ω.

The radiation field, which is treated classically, is composed of two counterpropagating monochromatic beams in a single spatial mode. The spatial dependence of Gaussian beams propagating in the $\pm z$ directions is, in cylindrical coordinates [12],

$$E_{\pm}(\rho,z)=E_0\frac{w_0}{w(z)}\exp\left(\pm i[kz+\eta(z)]-\rho^2\left[\frac{1}{w^2(z)}-\frac{\pm ik}{2R(z)}\right]\right), \tag{5}$$

where $\eta(z)=tn^{-1}(z/z_0)$, $w^2(z)=w_0^2[1+z^2/z_0^2]$, $R(z)=z(1+z_0^2/z^2)$. The beam divergence length is $z_0=\pi w_0^2/\lambda$, where w_0 is the beam waist at the focus $z=0$, and λ is the wavelength.

Assuming a linear polarization $\hat{\boldsymbol{\epsilon}}$ and dropping the counterrotating terms for absorption, Eqs. (4) and (5) yield

$$V(\mathbf{r},t)=-\frac{\boldsymbol{\mu}\cdot\boldsymbol{\epsilon}}{2}[E_+(\rho,z)+E_-(\rho,z)]e^{-i\omega t}. \tag{6}$$

III. TRANSITION AMPLITUDE

Standard second-order time-dependent perturbation theory yields the amplitude for the transition $|A,j,k,l\rangle\rightarrow|C,f,g,h\rangle$, through intermediate states $|B,\alpha,\beta,\gamma\rangle$:

$$C_{jkl}^{fgh}(t)=\sum_{b,\alpha,\beta,\gamma}\frac{1}{(i\hbar)^2}\int_0^t dt''\int_0^{t''}dt'\times\exp[i(\omega_{cb}+\Omega_{\alpha\beta\gamma}^{fgh})t'']\times\exp[i(\omega_{ba}+\Omega_{jkl}^{\alpha\beta\gamma})t']\times\langle h,g,f,C|V(\mathbf{r},t'')|B,\alpha,\beta,\gamma\rangle\times\langle\gamma,\beta,\alpha,B|V(\mathbf{r},t')|A,j,k,l\rangle. \tag{7}$$

Here $\Omega_{jkl}^{\alpha\beta\gamma}\equiv\Omega_j^{\alpha}+\Omega_k^{\beta}+\Omega_l^{\gamma}=\Omega[(\alpha-j)+(\beta-k)]+\Omega_z(\gamma-l)$ is the Bohr frequency associated with the transition between translational states $|j,k,l\rangle\rightarrow|\alpha,\beta,\gamma\rangle$. Evaluating the electronic dipole matrix elements we obtain

$$C_{jkl}^{fgh}(t)=\sum_{b,\alpha,\beta,\gamma}\frac{\mu_{ab}\mu_{bc}}{(i\hbar)^2}\int_0^t dt''\int_0^{t''}dt'\times\exp[i(\omega_{cb}+\Omega_{\alpha\beta\gamma}^{fgh}-\omega)t'']\times\exp[i(\omega_{ba}+\Omega_{jkl}^{\alpha\beta\gamma}-\omega)t']\times\frac{1}{4}\langle h,g,f|[E_+(\boldsymbol{\rho},z)+E_-(\boldsymbol{\rho},z)]|\alpha,\beta,\gamma\rangle\times\langle\gamma,\beta,\alpha|[E_+(\boldsymbol{\rho},z)+E_-(\boldsymbol{\rho},\gamma)]|j,k,l\rangle. \tag{8}$$

Assuming that the radiation fields are far off resonance from the intermediate state $|B\rangle$, contributions to $C_{jkl}^{fgh}(t)$ are important only when $t'\approx t''$ [13]. Consequently, we can substitute $\Omega_{\alpha}^{f}t''+\Omega_j^{\alpha}t'\approx\Omega_j^{f}t''$ and use the closure relation to eliminate the intermediate translational states, $|\alpha,\beta,\gamma\rangle$. For Doppler-free excitation, which involves the absorption of two counterpropagating photons, we disregard the products of fields from the same beam. The result is

$$C_{jkl}^{fgh}(t)=\sum_b\frac{\mu_{ab}\mu_{bc}}{2(i\hbar)^2}\int_0^t dt''\int_0^{t''}dt'\times\exp[i(\omega_{cb}+\Omega_j^f+\Omega_k^g+\Omega_h^l-\omega)t'']\times\exp[i(\omega_{ba}-\omega)t']\times\langle h,g,f|E_+(\rho,z)E_-(\rho,z)|j,k,l\rangle, \tag{9}$$

which yields the following expression for the transition amplitude:

$$C_{jkl}^{fgh}(z,t)=\sum_b\frac{\mu_{ab}\mu_{bc}}{(i\hbar)^2}\frac{1}{i(\omega_{ba}-\omega)}\times\frac{\exp[i(\omega_{ca}+\Omega_j^f+\Omega_k^g+\Omega_h^l-2\omega)t]-1}{i(\omega_{ca}+\Omega_j^f+\Omega_k^g+\Omega_h^l-2\omega)}\times\langle h_z|\left[\langle f|e^{-2x^2/w^2(z)}|j\rangle\times\frac{E_0^2w_0^2}{2w^2(z)}\langle g|e^{-2y^2/w^2(z)}|k\rangle\right]|l_z\rangle, \tag{10}$$

where $\omega_{ca}=(E_c-E_a)/\hbar$.

IV. LONG RAYLEIGH LENGTH LIMIT

For clarity it is convenient to consider the case in which the extent of the atomic cloud along the axis is much shorter than the laser beam divergence length. The general case is treated later. We substitute $w^2(z)\approx w_0^2$ and $\langle h_z|l_z\rangle=\delta_{h,l}$ in Eq. (10) to obtain

$$C_{jkl}^{fgl}=\sum_b\frac{\mu_{ab}\mu_{bc}}{(i\hbar)^2}\frac{E_{0_+}E_{0_-}}{2i(\omega_{ba}-\omega)}\times\frac{\exp[i(\omega_{ca}+\Omega_j^f+\Omega_k^g-2\omega)]-1}{i(\omega_{ca}+\Omega_j^f+\Omega_k^g-2\omega)}\times\langle f|e^{-2x^2/w_0^2}|j\rangle\langle g|e^{-2y^2/w_0^2}|k\rangle. \tag{11}$$

Photoexcitation involves simultaneous electronic and vibrational transitions, accompanied by a frequency shift due to the change in vibrational energy, as in a molecular transition involving change in electronic and vibrational levels. In

this case the ''molecule'' is the very soft atom-trap combined system. The line shape is governed by the translational matrix elements. We now turn to the physical processes that underlie these matrix elements, designated by

$$I_j^f(w_0) \equiv \langle f | e^{-2x^2/w_0^2} | j \rangle. \tag{12}$$

A. Momentum transfer and selection rules in two limiting cases

If $|j\rangle$ and $|f\rangle$ are momentum eigenstates with momenta p_j and p_f, respectively, Eq. (12), becomes

$$I_j^f(w_0) \propto \int dx e^{-i(p_j - p_f)x/\hbar} e^{-2x^2/w_0^2}. \tag{13}$$

It is evident that $I_j^f(w_0)$ is the spatial Fourier transform of the beam profile at momentum $p_j - p_f$. Consequently, the photoexcitation process effectively decomposes the laser beam profile into its momenta components. Each component contributes to absorption at a frequency shifted from the electronic transition frequency ω_{ca} by an amount Ω_j^f. If the translational energy is not quantized, the translational wave functions form a continuum and the continuum of frequency shifts produces a single broad line.

Atoms spatially confined, however, absorb in a discrete series of lines. We can extract the selection rules for the transitions in Eq. (12) by simple parity conservation. The Gaussian function in the matrix element I_j^f has even parity which for $I_j^f \neq 0$ requires that $|f\rangle$ and $|j\rangle$ have the same parity. This means that $f+j$ must be even. This rule follows from the symmetry of the system and would be violated if, for instance, the laser beam were not centered at the trap axis.

In the limiting case where the laser beam waist w_0 is much larger than the physical extent of either $|j\rangle$ or $|f\rangle$, i.e., $w_0 \gg \sqrt{(f,j)+1/2}/\alpha$, the Gaussian function in Eq. (12) is approximately constant and equal to unity. In this case,

$$I_j^f(w_0) = \langle f | j \rangle = \delta(j,f). \tag{14}$$

Because there is no change in the vibrational state, no sidebands arise from this transition and transit time broadening is absent. The situation is an extreme case of Dicke narrowing, in which motional broadening is suppressed when an atom is confined and cannot experience the spatial phase change of the electric field.

In the opposite limit where the beam waist is much smaller than the spatial extent of the wave functions, the Gaussian factor behaves like a normalized δ function. So, for an atom initially in state $|j\rangle$, the number of possible transitions to different final states $|f\rangle$ is enormous. The transition frequencies are all separated by an interval of 2Ω. The result is a fine comblike structure of lines within a broad envelope whose shape will be calculated below.

These two limiting cases will be shown to occur naturally in the general solution to Eq. (12) that we now discuss.

V. GENERAL BEHAVIOR

The exact solution to Eq. (12) is presented in Appendix A. However, because that solution is not transparent, particularly for large quantum numbers, we shall employ an approximate solution. The details of this approximation are presented in Appendix B. The key idea is that a harmonic trap potential is almost flat at the origin so that near the origin its eigenfunctions behave much like those of a free particle. As explained in Appendix B, the translational matrix elements in Eq. (10) can then be written

$$\langle f | e^{-2x^2/w^2(z)} | j \rangle \approx \frac{\alpha w(z) \exp[-\alpha^2 w^2(z)/8(\sqrt{2j+1} - \sqrt{2f+1})^2]}{[4\pi^2(2j+1)(2f+1)]^{1/4}}. \tag{15}$$

The structure of the y matrix element is identical and parity considerations again dictate that the matrix element vanishes unless $f+j$ is even. We shall postpone treatment of the longitudinal matrix elements, leaving them in terms of the initial and final longitudinal quantum numbers l_z and h_z, respectively. Substituting Eq. (15) into Eq. (10), and using Eq. (5), we obtain

$$C_{jkl}^{fgh}(t) = \frac{E_0^2}{2(i\hbar)^2} \left[\sum_b \frac{\mu_{ab}\mu_{bc}}{i(\omega_{ba} - \omega)} \right] \times \frac{\exp[i(\omega_{ca} + \Omega_j^f + \Omega_k^g + \Omega_l^h - 2\omega)t] - 1}{i(\omega_{ca} + \Omega_j^f + \Omega_k^g + \Omega_l^h - 2\omega)} \times \frac{R^2 \langle h_z | \exp[-(1 + z^2/z_0^2)\psi] | l_z \rangle}{2\pi[(2j+1)(2f+1)(2k+1)(2g+1)]^{1/4}}, \tag{16}$$

where

$$\psi \equiv \frac{R^2}{8}[(\sqrt{2j+1} - \sqrt{2f+1})^2 + (\sqrt{2k+1} - \sqrt{2g+1})^2] \tag{17}$$

and $R = \alpha w_o$.

The parameter R, the ratio of the radii of the laser beam and the trap ground-state diameter, characterizes the qualitative behavior of the spectrum.

In the long Rayleigh length limit where z_o is large compared to the longitudinal extent of the trapped atoms, we can take $\langle h_z | \exp[-(1+z^2/z_0^2)\psi] | l_z \rangle \approx \exp[-\psi] \delta(h_z, l_z)$. Even if this condition is not satisfied, z_0 will usually be long compared to the spatial oscillation of the axial wave functions. Consequently, transitions between longitudinal states will not be significant. Alternatively, we can say that in the Doppler-free configuration the momentum transfer along the axis of the beam is negligible. Consequently, the longitudinal matrix elements take the form

$$I_z(l) = \langle l_z | \exp[-(1 + z^2/z_0^2)\psi] | l_z \rangle. \tag{18}$$

Assuming that the envelope of $|l_z\rangle$ is constant over the longitudinal extent of the trap state, $(\pm\sqrt{l+1/2}/\alpha_z)$, the result is

$$I_z(l)=\left[\frac{\sqrt{\pi}}{2}\frac{\mathrm{erf}(\zeta_l)}{\zeta_l}\right]\exp[-\psi], \tag{19}$$

where

$$\zeta_l\equiv\frac{\sqrt{\psi(l+1/2)}}{\alpha_z z_0}, \tag{20}$$

and erf() is the error function. If the atoms are confined in a distance small compared to the laser divergence length z_0, then $\zeta_l\to 0$. In the opposite limit $\zeta_l\to\infty$. The expression in brackets in Eq. (19) has the expected limiting behavior

$$\frac{\sqrt{\pi}}{2}\frac{\mathrm{erf}(\zeta_l)}{\zeta_l}\to 1 \quad \text{as} \quad \zeta_l\to 0, \tag{21}$$

$$\frac{\sqrt{\pi}}{2}\frac{\mathrm{erf}(\zeta_l)}{\zeta_l}\to 1/\zeta_l \quad \text{as} \quad \zeta_l\to\infty. \tag{22}$$

Substituting Eq. (19) into Eq. (16), we obtain

$$\begin{aligned}C_{jkl}^{fgl}(t)=&\frac{E_0^2}{2(i\hbar)^2}\left[\sum_b\frac{\mu_{ab}\mu_{bc}}{i(\omega_{ba}-\omega)}\right]\\&\times\frac{(\alpha w_0)^2\exp[-\psi]}{2\pi[(2j+1)(2f+1)(2k+1)(2g+1)]^{1/4}}\\&\times\frac{\sqrt{\pi}}{2}\frac{\mathrm{erf}(\zeta_l)}{\zeta_l}\frac{\exp[i(\omega_{ca}+\Omega_j^f+\Omega_k^g-2\omega)t]-1}{i(\omega_{ca}+\Omega_j^f+\Omega_k^g-2\omega)}.\end{aligned} \tag{23}$$

The population of level $|C\rangle$ evolves as

$$|C(t)|^2=\sum_{jfkgl}|C_{jkl}^{fgl}(t)|^2|A_{jkl}|^2, \tag{24}$$

where A_{jkl} is the initial population in state $|A,j,k,l\rangle$.

For a time long compared to the inverse of the natural (or homogeneously broadened) linewidth, the time dependence of Eq. (24) behaves as $2\pi t\delta(\omega_{ca}+\Omega_j^f+\Omega_k^g-2\omega)$. In this case we can define a transition rate $W_{jkl}^{fgl}=d|C_{jkl}^{fgl}(t)|^2/dt$. We introduce a phenomenological linewidth due to spontaneous emission, interactions, or other sources of homogeneous broadening. This line shape is determined by a distribution of ω_{ca} with full width at half-maximum Γ_c, and can be described by a normalized line-shape function $g_{ajk}^{cfg}(\omega)$, centered when $2\omega=\omega_{ca}+\Omega_j^f+\Omega_k^g$. Integrating the δ function convolved with $g_{ajk}^{cfg}(\omega)$, we obtain for the transition rate

$$\begin{aligned}W_{jkl}^{fgl}=&\left|\sum_b\frac{\mu_{ab}\mu_{bc}}{i(\omega_{ba}-\omega)}\right|^2\left|\frac{E_0^2}{2(i\hbar)^2}\right|^2\\&\times\left(\frac{\alpha^2 w_0^2 e^{-\psi}}{2\pi[(2j+1)(2f+1)(2k+1)(2g+1)]^{1/4}}\right)^2\\&\times\left(\frac{\sqrt{\pi}}{2}\frac{\mathrm{erf}(\zeta_l)}{\zeta_l}\right)^2 2\pi g_{ajk}^{cfg}(\omega).\end{aligned} \tag{25}$$

VI. LINE SHAPES

The line shape is given by Eq. (25). In the following cases, we will assume that the beam divergence length is longer than the sample, the limiting case described by Eq. (21).

A. Classical result

Biraben *et al.* [14] analyzed two-photon excitation treating the center-of-mass motion classically. They found that an atom moving transversely through the laser beam with velocity v_r contributes to absorption at a detuning $\delta=2\omega-\omega_{ca}$ according to

$$W(\omega)\propto\frac{E_o^4}{v_r^2}\int_{-\infty}^{+\infty}d\zeta\frac{\Gamma_c}{(\zeta-\delta)^2+\Gamma_c^2/4}\exp\left(-\frac{\zeta^2}{4v_r^2/w_0^2}\right). \tag{26}$$

We recover this result as follows. If in Eq. (25) the quantum numbers contained in ψ [Eq. (17)] are large, we can substitute $(\sqrt{2j+1}-\sqrt{2f+1})^2\approx n^2/(2j+1)$, where $n=f-j$. Expressing the translational energy in terms of velocity, we substitute $(2j+1)\to mv^2/(\hbar\Omega)=\alpha^2 v_x^2/\Omega^2$. We then sum Eq. (25) for all transitions starting from state j and finishing at state $j+n$ (keeping the y dimension constant), with a normalized line shape function $g_{Aj}^{C(j+n)}$,

$$W(\omega)\propto\left(\frac{E_0^2}{v_x}\right)^2\sum_n g_{aj}^{c(j+n)}\exp\left(-\frac{(n\Omega)^2}{4v_x^2/w_0^2}\right). \tag{27}$$

Taking the line shape to be Lorentzian, we substitute $g_{Aj}^{C(j+n)}=\Gamma_C/[(n\Omega-\delta)^2-\Gamma_C^2/4]$ and obtain

$$W(\omega)\propto\left(\frac{E_0^2}{v_x}\right)^2\sum_n\frac{\Gamma_c}{(n\Omega-\delta)^2-\Gamma_c^2/4}\exp\left(-\frac{(n\Omega)^2}{4v_x^2/w_0^2}\right), \tag{28}$$

which is essentially the same result as Eq. (26) if one treats $n\Omega$ as a continuous variable, and we substitute the sum for an integral. As Biraben *et al.* show [14], in a sample at temperature T with a Maxwell-Boltzmann distribution of momentum, this leads to an exponential line shape [see Fig. 1(a)], $W(\delta\omega)\propto\exp[-|\delta\omega|/\Delta\omega]$, where $\delta\omega=2\omega-\omega_{ca}$ and $\Delta\omega=\sqrt{2kT/m}/w_o$.

B. Trapped atom spectrum

Returning to the case of trapped atoms, for a thermalized sample we must sum over all possible sets of (j,k,l) weighted by the initial population distribution, given by the appropriate statistics. The spectrum can be computed by calculating the one-dimensional spectrum, considering only the x or y motion, and convolving it with itself to generate the spectrum, i.e.,

$$W_{2d}(d\omega)=\sum_{d\omega_x}W_{1d}(d\omega_x)W_{1d}(d\omega-d\omega_x). \tag{29}$$

The resulting spectra are shown in Fig. 1. From (a) to (c) the trap oscillation frequency increases with respect to the time-of-flight linewidth, resulting in fewer sidebands in the

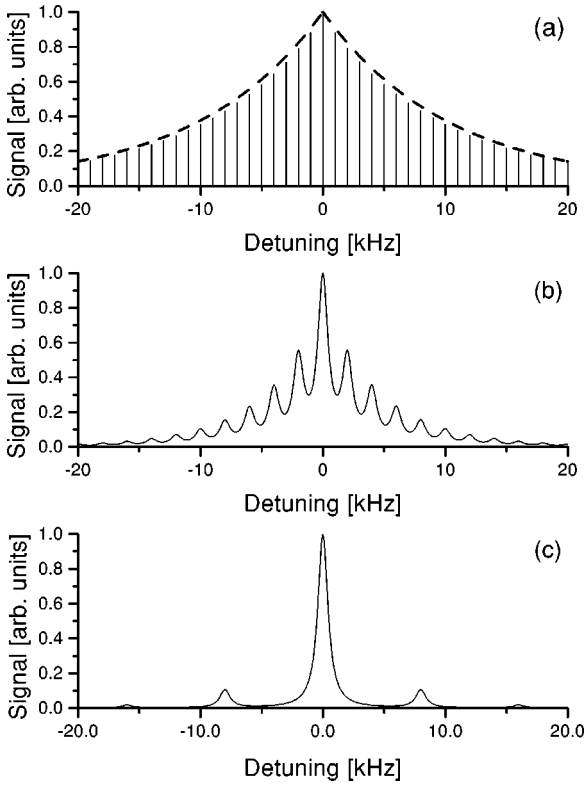

FIG. 1. Calculated spectra for various values of laser linewidth γ_c, trap frequency Ω, laser beam waist w_0, and temperature $T=100\ \mu$K. (a) Picket fence spectrum from quantized motion and semiclassical time-of-flight envelope in dashed line: $\gamma_c \to 0$, $\Omega=2\pi\times 0.5$ kHz, $w_0=20\ \mu$m; (b) $\gamma_c=2\pi\times 1$ kHz, $\Omega=2\pi\times 1$ kHz, $w_0=40\ \mu$m; (c) same as (b) but $\Omega=2\pi\times 4$ kHz.

spectrum. Spectrum (a) shows, in vertical bars, the relevant transitions in the limit of $\Gamma_c \to 0$, with the semiclassical exponential line shape as an envelope. The semiclassical line shape is the result of this quantized calculation in the limit where the trap frequency tends to zero. Different values of Γ_c (intrinsic resolution) are shown in (b). When the trap oscillation frequency is large compared to the intrinsic spectral resolution, the spectrum clearly resolves the sidebands allowing a much higher resolution than possible with time-of-flight broadening. The ratio Ω/Γ_c sets the contrast of the fringes which still lie under the exponential line shape.

As discussed in Sec. IV, these sidebands result from simultaneous electronic and vibrational transitions. Alternatively, one can understand these as Ramsey's fringes in a multipassages configuration. As the atom transverses the field many times without losing coherence, the spectrum contains interference fringes.

It is to be noted that the narrow line shape does not result from a velocity-selective process, as, for instance, in saturated absorption. Therefore, this process typically results in an increased signal strength. For example, the spectrum in Fig. 1(c), will have a much higher peak and signal-to-noise ratio than the equivalent one in (b), with the only change being a higher trap frequency.

If the trap were not harmonic, i.e., with nonuniform energy level separations, the peaked structure could completely wash out. Another effect that could easily change this result is a residual misalignment between the beams, since it would add some extra momentum component in the radial direction besides the one provided by the variation of the intensity.

VII. CONCLUSION

By explicitly introducing the quantized states of a harmonic trap, we have developed expressions for the spectral line shape of trapped atoms interacting with a Gaussian laser beam by Doppler-free two-photon absorption, in the regime of low intensities. When the experimental broadening, excluding transit-time broadening, is less than the trap level energy separations, the spectrum displays resolved peaks that are separated by twice the trap oscillation frequency. These lie under the envelope of a typical transit-time broadened line shape. When the quantum features are not resolvable, our result recovers the semiclassical result [14].

We have reinterpreted the phenomenon of transit-time broadening in terms of transverse momenta redistribution from the laser beam to the atom. In this quantized picture, the atom only accepts momenta that transfer if from one eigenstate to another. This formalism can be extended in a straightforward manner to account for the change in resonance frequency with trapping field [15]. Such an effect exists for magnetically trapped spin-polarized hydrogen and antihydrogen, where it should play a considerable role in precision test of CPT violation [16]. The formalism presented here can be easily extended to different trap shapes as well as different laser beam profiles.

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APPENDIX A: SOLUTION TO EQ. (12)

In this section we present an exact solution to the equation

$$I_j^f(w_0) \equiv \langle f | e^{-2x^2/w_0^2} | j \rangle. \tag{A1}$$

The wave functions for the harmonic oscillator $|f\rangle, |j\rangle$ can be written in terms of Hermite polinomials $H_n()$ as

$$\Psi_j(\alpha x) = N_j e^{-\alpha^2 x^2/2} H_j(\alpha x), \tag{A2}$$

where $N_j \equiv 1/(2^j j!\,\pi^{1/2})^{1/2}$ is the normalization factor. By defining $\xi \equiv \alpha x$ and $\gamma^2 \equiv 2/(\alpha w_0)^2$, we can rewrite Eq. (A1) as

$$I_j^f = N_j N_f \int d\xi e^{-(1+\gamma^2)\xi^2} H_j(\xi) H_f(\xi). \tag{A3}$$

We can now make use of the generating function for the Hermite polynomials [17]:

$$g(\xi,t) = e^{-t^2+2t\xi} = \sum_{n=0}^{\infty} H_n(\xi) \frac{t^n}{n!}. \tag{A4}$$

Consider then the following relation:

$$\int d\xi \exp[-(1+\gamma^2)\xi^2] g(\xi,s) g(\xi,t) = \sqrt{\frac{\pi}{1+\gamma^2}} e^{[-t^2(\gamma^2/1+\gamma^2)]} e^{[-s^2(\gamma^2/1+\gamma^2)]} e^{[st(2/1+\gamma^2)]}. \tag{A5}$$

But this is also equal to

$$a = \sum_{n,m} \frac{t^n s^m}{n!m!} \int d\xi e^{-(1+\gamma^2)\xi^2} H_n(\xi) H_m(\xi) \equiv \sum_{n,m} \frac{t^n s^m}{n!m!} C_{n,m}.$$

If we expand the exponentials of s and t and equate the terms with identical powers in s and t for the equations above, we obtain

$$C_{n,n+2k} = \frac{n!(n+2k)!\sqrt{\pi}}{\sqrt{1+\gamma^2}} \left(\frac{(-1)\gamma^2}{1+\gamma^2}\right)^{n+k} \times \sum_{j=n,n-2,\ldots} \frac{(-2/\gamma^2)^j}{\left(\frac{n-j}{2}\right)! \left(\frac{n+2k-j}{2}\right)! j!}. \tag{A6}$$

Inserting the normalizing factors, we obtain the desired result:

$$\langle n | e^{-2x^2/w^2} | n+2k \rangle = \frac{C_{n,n+2k}}{[2^{2(n+k)} n!(n+2k)!\pi]^{1/2}}, \tag{A7}$$

where k and n are positive integers.

APPENDIX B: APPROXIMATE SOLUTION TO EQ. (12)

The expression in Eq. (A7) above becomes difficult to handle for large n due to the factorials, and it fails to make apparent the underlying physics in Eq. (12). It is thus useful to derive a simpler, approximate solution to Eq. (12) as follows.

In a two-photon experiment one usually focuses the laser beam tightly to obtain an increase in the local intensity. The beam waist is then likely to be much smaller than the typical sample radius. For the majority of the atoms in the sample, the change in potential energy as it traverses the laser beam is much smaller than its total energy. In this case, we can treat the trap potential, in the interaction region, as constant. The wave function near the origin, where the atom interacts with the laser, becomes a free-particle-like wave function. Still, we consider the quantized levels with energy spacing given by the trap potential.

In order to judge the validity of this plane-wave approximation, we consider a typical experimental situation in cold hydrogen spectroscopy. Consider a sample at 100 μK held in the magnetic trap with a ground-state wave function extending over 10 μm in radius. The wave function for an atom at the thermal energy kT ($n \approx 20\,000$) extends over 1600 μm in radius while a typical laser beam waist is 40 μm. Clearly the plane-wave approximation should be excellent for atoms with energies around kT, while it is expected to be poor for the lowest levels. In fact, we find that the approximate solution in Eq. (B6) differs from the exact values given by Eq. (A7) by 10% for $j=10 \rightarrow f=10$, and only 1.2% for $j=20 \rightarrow f=22$ ($E_j \approx kT/1000$).

All the energy of level ''l,'' $E_l = \hbar\Omega(l+1/2)$, is contained in the kinetic energy term $p^2/2m$ around $\rho=0$. Therefore, using the free-particle wave-function approximation we can write for the wave function of the ket $|l\rangle$ in the x representation

$$|l\rangle \rightarrow \frac{\alpha^{1/2} X_l}{2} [e^{i\alpha\sqrt{2l+1}x} + (-1)^l e^{-i\alpha\sqrt{2l+1}x}], \tag{B1}$$

where X_l is the peak amplitude of the wave function around the origin, soon to be determined.

Substituting this expression in Eq. (A1), we obtain

$$I_j^f(w_0) = \frac{\alpha^{1/2} X_j}{2} \frac{\alpha^{1/2} X_f}{2} \times \int dx [e^{i\alpha\sqrt{2j+1}x} + (-1)^l e^{-i\alpha\sqrt{2j+1}x}] \times e^{-2x^2/w_0^2} [e^{-i\alpha\sqrt{2f+1}x} + (-1)^l e^{i\alpha\sqrt{2f+1}x}]. \tag{B2}$$

The equation above reveals the same physics of momentum transfer as Eq. (13) when we used the free-particle wave function. Here we calculate the momentum components of the laser beam profile at wave vectors $(\pm\sqrt{2l+1} \pm \sqrt{2m+1})\alpha$. The result is

$$I_j^f(w_0) = \frac{X_j X_f \alpha w_0}{2[2/\pi]^{1/2}} \delta(j+f=\text{even}) [e^{-(\alpha w_0)^2/8(\sqrt{2j+1}-\sqrt{2f+1})^2} + (-1)^f e^{-(\alpha w_0)^2/8(\sqrt{2j+1}+\sqrt{2f+1})^2}]. \tag{B3}$$

For large quantum numbers, which is usually the case for a gas far from quantum degeneracy, we can neglect terms depending on the sum of the square roots.

In order to determine the coefficients X_j, X_f we resort to the power series of the Hermite polynomials in which case we can write for the even wave functions $\Psi_{2n}(x)$ [17]

$$\Psi_{2n}(0) = (-1)^n \frac{(2n)!}{2^n n!} \Big/ [(2n)!\pi^{1/2}]^{1/2}. \tag{B4}$$

To extend this result to odd wave functions, we approximate it by employing Stirling's approximation for the factorials and we obtain

$$X_j \approx \pm \left(\frac{2}{j\pi^2}\right)^{1/4}. \tag{B5}$$

Every time n changes by 2, the Hermite polynomials will change phase at the origin by π, and this sets the sign of $I_j^f \propto (-1)^{(j+f)/2}$. Since this expression will eventually be squared, we omit the sign. To extend the result to $n=0$ we add 1/2 to the quantum numbers in the denominator of Eq. (B5).

The final solution to Eq. (12) is then

$$I_j^f(w_0)=\frac{\alpha w_0 e^{-\alpha^2 w_0^2/8(\sqrt{2j+1}-\sqrt{2f+1})^2}}{[4\pi^2(2j+1)(2f+1)]^{1/4}}\delta(j+f=\text{even}). \tag{B6}$$

The same way one regards $\langle p_j\rangle_{\text{rms}}=\hbar\alpha\sqrt{2j+1}$ as a measure of momentum content of state $|j\rangle$ in a harmonic oscillator, we can also, inspecting the equation above, regard $\langle p_{\text{beam}}\rangle=\sqrt{2}\hbar/w_0$ as a measure of the momentum content of the laser intensity profile. We can easily interpret Eq. (B6) above by rewriting its exponential as $\exp[-2(\langle p_j\rangle_{\text{rms}}-\langle p_f\rangle_{\text{rms}})^2/p_{\text{beam}}^2]$. This expression has a clear physical interpretation: a transition $j\rightarrow f$ is only induced if the beam intensity profile carries enough momentum to account for the change of momentum in the transition.

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