I. INTRODUCTION

Excited-state transitions in atomic systems are finding an increasing range of applications including quantum information [1], optical filters [2], electric field sensing [3–5], and quantum optics [6,7]. They are also used for state lifetime measurements [8], frequency up-conversion [9], the search for new stable frequency references [10,11] and multiphoton laser cooling [12]. However, excited-state transitions are inherently more difficult to probe than ground-state transitions, especially if the lower state is short lived. It is possible to probe an excited-state transition directly if the dipole moment is strong enough [13], but more commonly excited-state character is observed by mapping onto ground-state transitions using electromagnetically induced transparency (EIT) in a ladder configuration. Using EIT it is possible to probe even relatively weak excited-state transitions, such as those to highly excited Rydberg states [14–16]. Nanosecond timescales have also been probed, effectively “freezing out” the motion of thermal atoms [17].

EIT involving Rydberg states has paved the way to recent advances in nonlinear and quantum optics [6] as the strong interactions among the Rydberg atoms lead to large optical nonlinearities, even at the single-photon level [7,18,19]. In room-temperature Rydberg gases, the atomic interactions can lead to a nonequilibrium phase transition [20], and evidence for strong van der Waals interactions has been observed [21]. Despite the considerable successes of ladder EIT, there is a particular class of energy level schemes for which ladder EIT cannot be observed in a Doppler-broadened medium. Specifically, when the upper transition wavelength is longer than the lower (“inverted-wavelength” system) [22,23], the transparency window is absent as it is smeared out by velocity averaging.

In this paper we make use of hyperfine quantum beats [24,25] to probe the excited-state transition dynamics of an inverted-wavelength ladder system in a thermal vapor. We find strong evidence for both Rabi oscillations and sub-Doppler Autler-Townes splitting.

The paper is organized as follows: In Sec. II we construct a toy model of our experiment, giving an overview of the physics involved. Section III details our experimental procedure and in Sec. IV we present results in both the time-domain and the frequency domain. Section V outlines a computer model that we developed to understand the signals, which we compare to the data in Sec. VI. The model yields good qualitative agreement, which allows us to interpret features that we observe in the frequency domain.

II. PRINCIPLE OF PERTURBED QUANTUM BEATS IN A LADDER SYSTEM

In this section we outline a toy model of our ladder system which includes the minimum possible complexity to illustrate the physical principle (Fig. 1). The toy model considers a zero-velocity atom with ground state $|g\rangle$, an intermediate excited state $|e\rangle$, and an upper excited state $|u\rangle$. There is also a reference state $|e'\rangle$ which is close in energy to $|e\rangle$. The transition from $|g\rangle \rightarrow |e\rangle$ is driven by a short pulse while a continuous wave (CW) laser drives the excited-state transition from $|e\rangle \rightarrow |u\rangle$. For a sufficiently short excitation pulse the bandwidth exceeds the energy interval between $|e\rangle$ and $|e'\rangle$, and a coherent superposition of the two states is prepared by the pulse. The dynamics of the excited-state transition are read out by measuring the fluorescence from states $|e\rangle$ and $|e'\rangle$.

We begin our explanation by considering the simple case when the excited-state transition driving field is switched off (left column of Fig. 1). Once the coherent superposition of states $|e\rangle$ and $|e'\rangle$ has been prepared, the total fluorescence decays exponentially according to the state lifetime. However, the fluorescence into an appropriately chosen mode, characterized by polarization and propagation direction, is modulated by beating [26]. These “quantum beats” represent interference between the two different quantum pathways associated with $|e\rangle$ and $|e'\rangle$. The interference is erased if information regarding which pathway was taken is recovered (e.g., spectroscopically resolving the fluorescence from each state). In our toy model
The level scheme (top), the Fourier spectrum, of the excited-state transition are written into the quantum beats. We split the decay rates of their energies. Right: Driving an excited-state transition prepared in a superposition of closely spaced excited states $\omega / \Omega_1$ driving of the excited-state transition it is easiest to consider the dressed-state picture well. 

The short pulse of light excites several states in the $6P_{1/2}$ manifold and a CW laser drives an excited-state transition $6P_{3/2} F = 5 \rightarrow 7S_{1/2} F = 4$. (b) Schematic of experiment: Vertically polarized beams counterpropagate through a cesium vapor cell and fluorescence from the $D_2$ transition is detected with a single-photon counter.

The simplified level scheme and experimental setup are shown in Figs. 2(a) and 2(b) respectively. We use cesium atoms in a vapor cell (length 2 mm) at room temperature (19°C). The laser intensity is most uniform, by virtue of tighter focusing of the excited-state transition driving field that the atoms experience. We excite the first transition using a short pulse (FWHM of 1 ns) of 852-nm light generated by a CW diode laser stabilized to the $F = 4 \rightarrow F' = 5$ hyperfine transition and modulated by a Pockels cell between two crossed, high-extinction polarizers. The short pulse duration means that the pulse intensity spans the hyperfine energy splitting of the $6P_{1/2}$ manifold and therefore prepares a coherent superposition of several hyperfine states. It is this coherent superposition of states that leads to quantum beats in our system. The excited-state transition is driven by a counterpropagating, CW laser beam locked to the $6P_{3/2} F' = 5 \rightarrow 7S_{1/2} F = 4$ (1469 nm) transition using excited-state polarization spectroscopy [27].

To best control the effects of driving the excited-state transition, it is desirable to minimize the spread of intensity of the excited-state transition driving field that the atoms experience. To achieve this, we only sample the center of the CW driving laser beam ($1/e^2$ radius 0.3 mm) where the intensity is most uniform, by virtue of tighter focusing of the preparation pulse ($1/e^2$ radius 0.06 mm). Both the laser beams are vertically polarized, and we detect fluorescence.
The dashed vertical lines correspond to the 6$^2$ hyperfine splitting relation to the unperturbed fluorescence. In panels (d) and (h) we show the spectrum of the difference between the two polarization signals. The spectra are normalized signals. The solid line (blue online) shows the vertically polarized fluorescence and the dashed line (red online) shows the horizontally polarized fluorescence. In panels (c) and (g) we present the Fourier spectra, calculated by taking the magnitude of the Fourier transform of the time-dependent fluorescence measurements of time-dependent vertically polarized fluorescence [(a) and (e)] and horizontally polarized fluorescence [(b) and (f)]. In Fig. 3(d) we remove frequency components relating to the exponential decay envelope by subtracting the two signals (see the Appendix for details of normalization and subtraction). Because the beating of the two polarization signals is out of phase we retain the quantum beat frequency components and so we observe peaks at 201, 251, and 452 MHz, corresponding to the $6P_{3/2}$ hyperfine splitting [28] (highlighted with vertical dashed lines). The peak relating to the $F' = 3 \rightarrow F' = 4$ quantum beat (201 MHz) is very weak as the population in these two states is limited. This restricted population is a result of both weaker coupling to the ground state and also detuning from the middle of the excitation pulse bandwidth which is centered on the $F = 4 \rightarrow F' = 5$ transition.

When we drive the excited-state transition the quantum beats are modified. For the driving field intensity at the center of the laser beam $I_d = 4 \text{ W cm}^{-2}$, we present the vertically and horizontally polarized fluorescence measurements in Figs. 3(e) and 3(f) respectively, along with their Fourier spectra in Fig. 3(g) and the spectrum of the difference signal in Fig. 3(h). We can see the changes to the Fourier spectra that we expected from considering the toy model in Sec. II. First the peak relating to the $F' = 5 \rightarrow F' = 4$ beat (251 MHz) is split in two [Figs. 3(g) and 3(h)]. The origin of this effect is Autler-Townes splitting of the $6P_{3/2}F' = 5$ atomic state, caused by driving the excited-state transition. Second a different oscillation appears, leading to a peak at 100 MHz in this example [Fig. 3(g)]. This represents atoms performing Rabi oscillations on the excited-state transition. The absence of this peak from the difference signal in Fig. 3(h) is because the Rabi oscillations modulate the entire 852-nm fluorescence. Therefore the oscillation is in phase between the vertically and horizontally polarized fluorescence and is removed in the difference signal.

It is interesting to note that while the simple model outlined in Sec. II predicts that the splitting of the beat frequency would be equal to the frequency of the Rabi oscillation, it is clear from

![FIG. 3. (Color online) Top row [(a)–(d)]: Measurements of fluorescence showing unperturbed hyperfine quantum beats. Bottom row [(e)–(h)]: Measurements of fluorescence showing quantum beats that are modified by CW driving field with intensity $I_d = 4 \text{ W cm}^{-2}$. We present measurements of time-dependent vertically polarized fluorescence [(a) and (e)] and horizontally polarized fluorescence [(b) and (f)]. In panels (c) and (g) we present the Fourier spectra, calculated by taking the magnitude of the Fourier transform of the time-dependent fluorescence signals. The solid line (blue online) shows the vertically polarized fluorescence and the dashed line (red online) shows the horizontally polarized fluorescence. In panels (d) and (h) we show the spectrum of the difference between the two polarization signals. The spectra are normalized such that the peak in the difference signal relating to the unperturbed $F' = 5 \rightarrow 4$ beat (d) has a height of 1 (see the Appendix for full details). The dashed vertical lines correspond to the $6P_{3/2}$ hyperfine splitting [28] and the shaded bands correspond to regions presented as color plots in Fig. 4.](image)
signal and the splitting of the shows the modulus of the Fourier transform of the difference increases in frequency with increasing laser power. Figure 4(b) diagonal feature corresponds to the Rabi oscillation, which of the vertically polarized fluorescence measurements. The relating to Autler-Townes splitting. Parts (a) and (b) relate to the oscillations. (b) The difference signal demonstrates a branched feature to the Rabi fluorescence shows a diagonal feature that corresponds to Rabi

Sec. VI using a comprehensive computer simulation outlined in Sec. VI. Finally we note that the higher frequency branch of pulses. Further consequences of this effect are discussed in Fig. 3(g) that this is not the case. The cause of this discrepancy stems from Doppler effects which we explore and explain in Sec. VI using a comprehensive computer simulation outlined in Sec. V.

In Fig. 4 we present color plots covering a range of excited-state transition laser driving intensities $I_0 = 0 \rightarrow 7$ W cm$^{-2}$, constructed from nine individual sets of intensity measurements. Figure 4(a) shows the modulus of the Fourier transform of the vertically polarized fluorescence measurements. The diagonal feature corresponds to the Rabi oscillation, which increases in frequency with increasing laser power. Figure 4(b) shows the modulus of the Fourier transform of the difference signal and the splitting of the $F' = 5 \rightarrow F' = 4$ beat into two separate branches is clear.

We also point out some further, more subtle effects. First, the fluorescence decays more slowly as the longer lived $7S_{1/2}F'' = 4$ state is mixed into the $6P_{3/2}$ states. Second, the total amount of measured 852-nm fluorescence decreases. This is partly because the atoms can now decay from the $7S_{1/2}F'' = 4$ state via the $6P_{1/2}$ manifold as well as the $6P_{3/2}$ manifold, but could also be due to hyperfine optical pumping caused by light leaking through the Pockels cell between pulses. Further consequences of this effect are discussed in Sec. VI. Finally we note that the higher frequency branch of the split $F' = 5 \rightarrow F' = 4$ quantum beats is stronger than the low frequency branch [Fig. 4(b)]. This effect is even more exaggerated in the $F' = 5 \rightarrow F' = 3$ (452 MHz) beat where we do not observe the low-frequency branch at all [Figs. 3(d) and 3(g)]. The absence of the lower branch originates from Doppler effects that we also discuss in Sec. VI.

V. COMPUTER MODEL

Here we develop a theoretical simulation to predict the behavior of our system. Conceptually, it involves two steps. First the optical Bloch equations for the system are solved numerically; second, the time-dependent expectation value of a “detection operator,” $\mathcal{B}$, is calculated, giving the expected fluorescence [26]. The operator has the form

$$\mathcal{B} = C \sum_f \mathbf{e} \cdot \hat{\mathbf{D}}(f) \langle f | e^* \cdot \hat{\mathbf{D}} \rangle,$$

where $C$ is a coefficient relating to detection efficiency, $\mathbf{e}$ is a unit vector describing the polarization of the detected fluorescence, and $\hat{\mathbf{D}}$ is the electric dipole operator for the $D_2$ transition of the atom. The final states $f$ include all of the magnetic sublevels ($m_F$) of the two $6S_{1/2}$ hyperfine ground states. We note that the expectation value of the operator $\mathcal{B}$ is proportional to the square of the atomic dipole projected onto the detected polarization angle and measures the coupling of the atomic state to the field modes. The calculation process is repeated for a sample of velocity classes, which are then summed and weighted according to a Boltzmann distribution.

The computation basis is fixed such that the linearly polarized excitation lasers drive only $\pi$ transitions, allowing our calculation to be performed in a set of mutually uncoupled $m_F$ subspaces. Note that this basis might not be the energy eigenbasis due to uncompensated laboratory magnetic fields. However, any coherence developed between the $m_F$ subspaces as a consequence of this can be neglected since the duration of our experiment is much shorter than the relevant Larmor precession time scale. The time evolution of the density matrix $\rho_{m_F}$ in each of the nine $m_F$ subspaces is calculated using a set of optical Bloch equations,

$$\dot{\rho}_{m_F} = \frac{i}{\hbar} [\rho_{m_F}, \hat{H}_{m_F}] - \Gamma,$$

where $\hat{H}_{m_F}$ is the Hamiltonian for each subspace and $\Gamma$ is a decay operator. The Rabi frequencies are calculated individually for each $m_F$ subspace and each subspace includes one state from each of the hyperfine levels: $6S_{1/2}F = 3, 4; 6P_{1/2}F' = 3, 4, 5,$ and $7S_{1/2}F'' = 4$. This convenient sub-division offers a computational speed up that permits the simulation to be run on a desktop computer.

Although the subspaces are not coupled by the driving laser fields, we note that they are not truly separate, since atoms can undergo spontaneous $\sigma^\pm$ transitions, resulting in a change of $m_F$ quantum number. Instead of modeling this full behavior, we attribute the total rate of spontaneous decay of each state to $\pi$ transitions, thus conserving the total population in each subspace. In this way, we are able to capture the lifetimes of the states and retain computational efficiency. Furthermore, because the time scale of our experiment is set by a single atomic state lifetime, we are confident that the effect of these angular momentum changing processes is negligible, as there is insufficient time to redistribute atomic population amongst the $m_F$ subspace.

In the final step of our model, we collate the populations and coherences from the nine subspaces into a single density matrix, giving the complete state of the atom as it changes in time. Using the “detection operator,” we project the atomic
dipole at each time step and hence infer both the linear and circularly polarized 852-nm fluorescence.

VI. ANALYSIS

In this section we make a direct comparison between the computer simulation and the measured data. In Fig. 5 we present the results of the vertically polarized fluorescence for both the experiment and simulation. The unperturbed hyperfine quantum beat signal fits well, and we see at least qualitative agreement for the perturbed beats. Although the features are often more pronounced in the simulation than the data, there is a qualitative match between the data and theory. On the strength of this we can draw additional physical insight about the system.

In Sec. IV we noted that the splitting of the $F' = 5 \rightarrow F' = 4$ hyperfine quantum beat was unexpectedly smaller than the measured frequency of the Rabi oscillation. We suggest this is similar to narrowed EIT windows in thermal vapors [30,31], where off-resonant velocity classes partially fill the transparency window left by resonant atoms. Calculated contributions to the splitting of the $F' = 4 \rightarrow F' = 5$ quantum beat from different velocity classes are shown in Fig. 6. The zero-velocity class (bold, green [gray]) shows a splitting that is consistent with the simulated excited-state transition Rabi frequency, yet this is much larger than the splitting which appears in the total signal. Figure 6 shows how contributions from off-resonant velocity classes fill in the gap. The inset compares this best-fit-calculated spectrum with our data and we see qualitative agreement, although we acknowledge a significant discrepancy in the simulated ($I_{\text{sim}} = 0.9$ W cm$^{-2}$) and measured ($I_d = 3$ W cm$^{-2}$) excited-state transition laser intensities.

We also noted in Sec. IV that the high-frequency branch of the split $F' = 4 \rightarrow F' = 5$ quantum beat makes a stronger contribution to the Fourier spectrum than the low-frequency branch. This unexpected asymmetry can be explained by constructing a two-step argument: First, we note that the strongest beats arise from atoms experiencing a red Doppler shift of the 852-nm laser. This moves the center of the frequency profile of the pulse between the two beating transitions, promoting the excitation of both levels as required for quantum beats. Second, the atoms which experience a red shift for the 852-nm laser see a blue shift of the 1469-nm laser because the laser beams are counterpropagating. This blue shift means that the dressed states represented in the higher frequency branch of the split quantum beat have a greater

FIG. 5. (Color online) Vertically polarized fluorescence: We compare the model (black line) and experimental data points (blue [gray]) for measured excited-state transition laser intensity $I_d = (0, 3, 6, 7)$ W cm$^{-2}$ (top to bottom). We note that the visibility of the peaks is always smaller than the model predicts. The incident light pulse occurs at time $t = 0$ ns and the error bars are calculated from Poissonian photon counting statistics [29].

FIG. 6. (Color online) Calculated magnitude of the normalized Fourier transform of the vertically polarized fluorescence: We show a breakdown of velocity class contributions, with dashed (red online) lines showing individual velocity classes traveling away from the 852-nm laser and solid (blue online) lines showing velocity classes traveling towards the 852-nm laser. The velocity classes are spaced at 33 m s$^{-1}$ intervals and the bold (green online) line shows the contribution from zero-velocity atoms. The gray shaded area shows the scaled sum of the signals and the calculation relates to probing a region with a uniform excited-state transition driving field intensity $I_{\text{sim}} = 0.9$ W cm$^{-2}$. The inset compares the data taken for measured CW driving field intensity $I_d = 3$ W cm$^{-2}$ shown in bold (blue online) and the summed model (gray shaded area and black line).
We have demonstrated a method using hyperfine quantum beat spectroscopy for observing sub-Doppler Autler-Townes-type splitting in an “inverted wavelength” ladder scheme which would not be observable in a continuously excited room-temperature vapor. A comprehensive model of the fluorescence gives qualitative agreement with our data, and we use it to gain physical insight into the process. By exploiting our model to its full potential it would be possible to combine information from both the Autler-Townes splitting and the Rabi oscillations to achieve a complete readout of excited-state transition dynamics. Our work on ladder excitation schemes contributes to a general effort towards the exploitation of Rydberg atoms in a room-temperature atomic vapor using multiphoton, stepwise excitation. In a wider context, our method offers a means for investigating excited-state transitions in a room-temperature vapor.

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APPENDIX: NORMALIZATION OF FOURIER SPECTRA

In Fig. 3 we present normalized Fourier spectra of the time domain signals. Here we inform the reader of the details of the normalization.

We begin with the measured time domain quantities. For unperturbed beats, \( h_0(t) \) and \( v_0(t) \) correspond to the horizontally and vertically polarized fluorescence respectively, and for the modified quantum beats \( h_m(t) \) and \( v_m(t) \) correspond to the horizontally and vertically polarized fluorescence respectively. These quantities are Fourier transformed to give \( H_0(\omega) \), \( V_0(\omega) \), \( H_m(\omega) \), and \( V_m(\omega) \). The horizontal and vertical signals are scaled and subtracted to give the difference signals, \( D_0(\omega) \) and \( D_m(\omega) \):

\[
D_0(\omega) = H_0(\omega) - \sum \frac{h_0(t)}{v_0(t)} V_0(\omega) \tag{A1}
\]

and

\[
D_m(\omega) = H_m(\omega) - \sum \frac{h_m(t)}{v_m(t)} V_m(\omega), \tag{A2}
\]

where \( D_0(\omega) \) and \( D_m(\omega) \) correspond to unperturbed beats and modified beats respectively. Scaling the vertically polarized fluorescence signal in this way means that the average of the difference signals is zero in the time domain.

A global normalization factor, \( n \), is found using \( D_0 \), corresponding to the height of the \( F' = 4 \rightarrow 5 \) (251 MHz) unperturbed quantum beat:

\[
n = |D_0(\omega_{F' = 4 \rightarrow 5})|, \tag{A3}
\]

Finally we plot the quantities \( H_0(\omega) \), \( V_0'(\omega) \), \( D_0'(\omega) \), \( H_m'(\omega) \), \( V_m'(\omega) \), and \( D_m'(\omega) \), corresponding to the normalized modulus of the Fourier spectra, such that

\[
H_0'(\omega) = |H_0(\omega)|/n, \nonumber \\
V_0'(\omega) = |V_0(\omega)|/n, \nonumber \\
D_0'(\omega) = |D_0(\omega)|/n, \nonumber \\
H_m'(\omega) = |H_m(\omega)|/n, \nonumber \\
V_m'(\omega) = |V_m(\omega)|/n, \nonumber \\
D_m'(\omega) = |D_m(\omega)|/n. \tag{A4}
\]

As a result of this normalization, the peak in the difference signal relating to the unperturbed \( F' = 5 \rightarrow 4 \) beat has a height of 1, and the results from different excited-state transition driving field strengths are directly comparable.

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