Structural and dynamical aspects of avoided-crossing resonances in a three-level Λ system

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In a recent publication [Phys. Rev. A **79**, 065602 (2009)], it was shown that an avoided-crossing resonance can be defined according to level-structural or dynamical criteria. We propose an experiment to observe the difference between the two definitions in a three-level Λ system using microwave fields coupling hyperfine magnetic sublevels in alkali-metal atoms.

I. INTRODUCTION

The concept of "resonance" is ubiquitous in physics. A resonance implies variations of different characteristic properties with respect to one parameter within the resonance width, but the extremal points for the different properties do not necessarily coincide. In a recent publication [1], we studied the definition of an avoided-crossing resonance in quantum systems with discrete energy levels. The parameter values of minimal splitting and of maximal transition probability do not coincide in general. In this brief paper, we propose a simple physical setting, i.e., a three-level system subjected to a two-photon transition, where the difference between structural and dynamical aspects of the resonance may be observed.

II. THE MODEL

Consider a three-level system in a Λ configuration (a Raman two-photon setup, as seen in Fig. 1) that is described, in a laser-adapted interaction picture, by the time independent Hamiltonian ($\hbar = 1$) [2]

$$H = \frac{1}{2}\begin{pmatrix} 0 & \Omega_1 & 0 \\ \Omega_1 & -2\delta_1 & \Omega_2 \\ 0 & \Omega_2 & -2(\delta_1 - \delta_2) \end{pmatrix}, \qquad (1)$$

where Ω_1 and Ω_2 are the Rabi frequencies of the transitions and δ_1 and δ_2 are the detunings as shown in Fig. 1. When the lasers are turned off ($\Omega_1 = \Omega_2 = 0$), the atomic states are uncoupled and the energy levels of H cross each other at $\delta_1 = 0$ and $\delta_1 = \delta_2$. When the coupling lasers are turned on, these crossings become avoided crossings, and transitions between the atomic energy levels at each resonance may occur (see Fig. 2).

An analytical diagonalization of the full Hamiltonian (1) is possible, but hardly illuminating. To have simple formulas and gain some understanding, approximations will be useful. Among the two resonances, we shall focus on the one at $\delta_1 = \delta_2$. The distance between them is δ_2, and since the splitting of each avoided crossing is proportional to the Rabi frequencies Ω_1 and Ω_2, the avoided crossings will be well isolated, leading to clean transitions, as long as $\delta_1 \sim \delta_2 \gg \Omega_1, \Omega_2$. Under this condition, the state $|2\rangle$ is scarcely populated

and can be adiabatically eliminated to give an effective two-level Hamiltonian

$$H_{\text{eff}} = \begin{pmatrix} -\delta_{\text{eff}} & \Omega_{\text{eff}} \\ \Omega_{\text{eff}} & \delta_{\text{eff}} \end{pmatrix}, \qquad (2)$$

which corresponds to an effective coupling between states $|1\rangle$ and $|3\rangle$ with effective coupling strength Ω_{eff} and effective detuning δ_{eff}:

$$\Omega_{\text{eff}} = \frac{\Omega_1 \Omega_2}{4\delta_1}, \quad \delta_{\text{eff}} = \frac{1}{2}(\delta_2 - \delta_1) + \frac{\Omega_2^2 - \Omega_1^2}{8\delta_1}. \qquad (3)$$

As described in [1], when the diagonal and nondiagonal terms in a two-dimensional Hamiltonian depend on the same parameter (δ_1 in this case), the location of the resonance is not uniquely defined.

III. STRUCTURAL AND DYNAMICAL DEFINITIONS

From a "structural" perspective of the energy-level diagram, the resonance may be defined as the point where the distance between the two branches of the avoided crossing is minimum, which is given by the condition

$$\frac{\partial}{\partial \delta_1} \sqrt{\delta_{\text{eff}}^2 + \left(\frac{\Omega_1 \Omega_2}{4\delta_1}\right)^2} = 0. \qquad (4)$$

For the condition $\delta_1 \sim \delta_2 \gg \Omega_1, \Omega_2$, the solution to the this equation is approximately given by (up to fourth-order terms in the frequencies)

$$(\delta_1)_S \approx \delta_2 + \frac{\Omega_2^2 - \Omega_1^2}{4\delta_2} - \frac{(\Omega_2^2 - \Omega_1^2)^2}{16\delta_2^3} + \frac{\Omega_1^2 \Omega_2^2}{4\delta_2^3}. \qquad (5)$$

From a "dynamical" perspective, the resonance is defined by the value of δ_1 for which the transition probability from state $|1\rangle$ to state $|3\rangle$ is maximum. Using H_{eff}, this probability is easily computed as

$$P_{13} = \frac{\Omega_{\text{eff}}^2}{\delta_{\text{eff}}^2 + \Omega_{\text{eff}}^2} \sin^2\left(t\sqrt{\delta_{\text{eff}}^2 + \Omega_{\text{eff}}^2}\right) \qquad (6)$$
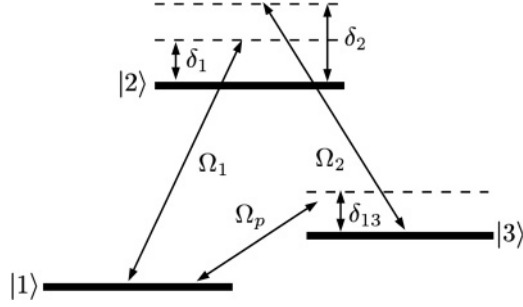
FIG. 1. Simple Raman three-level setup with energy levels $|1\rangle$, $|2\rangle$, and $|3\rangle$, and detunings of the two lasers with respect to atomic transitions and coupling strengths (Rabi frequencies) Ω_1 and Ω_2. Initially, states $|1\rangle$ and $|3\rangle$ are assumed to be uncoupled, where $\Omega_p = 0$. For the experimental determination of the different shifts, a coupling between states $|1\rangle$ and $|3\rangle$ becomes necessary, where $\Omega_p \neq 0$.

and shows a maximum at $\delta_{\text{eff}} = 0$, which corresponds to

$$(\delta_1)_D = \tfrac{1}{2}\left(\delta_2 + \sqrt{\delta_2^2 + \Omega_2^2 - \Omega_1^2}\right) \tag{7}$$

$$\approx \delta_2 + \frac{\Omega_2^2 - \Omega_1^2}{4\delta_2} - \frac{\left(\Omega_2^2 - \Omega_1^2\right)^2}{16\delta_2^3}. \tag{8}$$

The expressions (5) and (8) are separated by a dynamical shift Δ_D given in this approximation by

$$\Delta_D = (\delta_1)_S - (\delta_1)_D \approx \frac{\Omega_1^2 \Omega_2^2}{4\delta_2^3}, \tag{9}$$

which is plotted in Fig. 3 versus the ratio between the Rabi frequencies. There is good agreement between the exact dynamical shift and the approximation (9) for weak couplings. For strong couplings, the perturbative approach breaks down, but Eq. (9) still gives a good estimate.

IV. EXPERIMENTAL DETERMINATION

The "dynamical resonance" in Eq. (8) can be determined experimentally by preparing the system in state $|1\rangle$ for each δ_1
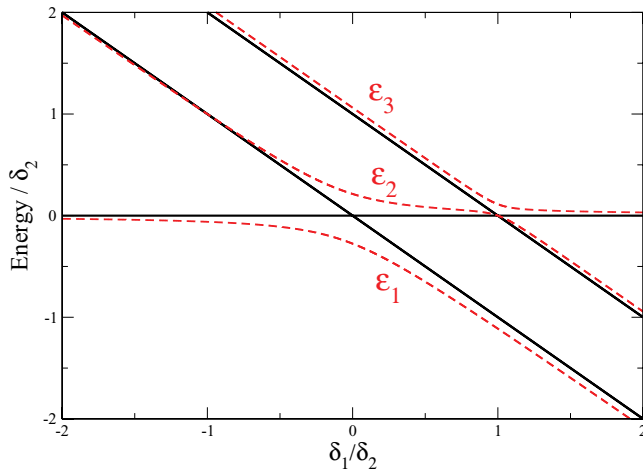

FIG. 2. (Color online) Bare (black solid line) and dressed (red dashed line, $\Omega_1 = \Omega_2 = 0.5\delta_2$) energy levels as a function of δ_1/δ_2.
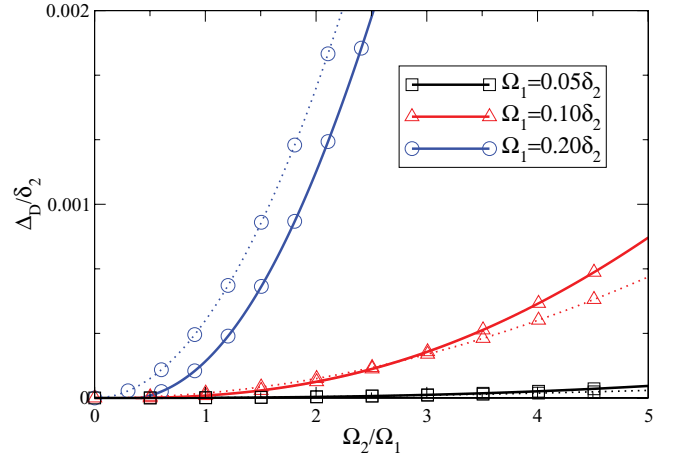

FIG. 3. (Color online) Exact dynamical shift computed by numerically diagonalizing the full three-level Hamiltonian (1) (solid lines) as a function of the ratio between the coupling strengths. The approximate expression for the dynamical shift (9) is plotted with dotted lines.

and looking for the maximum probability of finding state $|3\rangle$. The experimental determination of the minimum level splitting ("structural resonance") requires more work. One way is to use a third, auxiliary weak probe field connecting states $|1\rangle$ and $|3\rangle$, as in Fig. 1 with $\Omega_p \neq 0$. This transition may be electric-dipole forbidden, such as a magnetic-dipole-allowed transition, which is usually much weaker than electric-dipole transitions, a good thing in this context since we are interested in probing the dressed energy levels without excessively perturbing the original system.

The Hamiltonian describing the full system (including the probe field) takes the time-dependent form $H(t) = H + W(t)$, since, in general, there is no interaction picture in which the full Hamiltonian is time independent. Here, H is the Hamiltonian of the original system already given in Eq. (1), and the time-dependent perturbation is given by $W(t) = \frac{\Omega_p}{2}(|3\rangle\langle 1|e^{i\nu t} + \text{H.c.})$, where $\nu = \delta_1 - \delta_2 - \delta_{13}$, and δ_{13} is the detuning of the probe field with the $|1\rangle \leftrightarrow |3\rangle$ transition (see Fig. 1).

We shall examine hereafter the resonance at $\delta_1 \approx \delta_2$ (see Fig. 2). The dressed states will be labeled with increasing energy ($\epsilon_1 < \epsilon_2 < \epsilon_3$), so we have to measure the energy difference between ϵ_3 and ϵ_2 for determining the structural resonance.

We shall now consider H as a zeroth-order Hamiltonian weakly perturbed by $W(t)$, and use time-dependent perturbation theory to obtain the transition rate from dressed state $|\epsilon_2\rangle$ to dressed state $|\epsilon_3\rangle$:

$$
\begin{aligned}
P_{|\epsilon_2\rangle \to |\epsilon_3\rangle} &= \left| -i \int_0^t dt' \langle \epsilon_3 | W(t') | \epsilon_2 \rangle e^{i(\epsilon_3 - \epsilon_2)t'} \right|^2 \\
&= \Omega_p^2 \left[\frac{\alpha_{31}^2 \sin^2 \frac{(\Delta\epsilon + \nu)t}{2}}{(\Delta\epsilon + \nu)^2} + \frac{\alpha_{13}^2 \sin^2 \frac{(\Delta\epsilon - \nu)t}{2}}{(\Delta\epsilon - \nu)^2} \right. \\
&\quad \left. + \frac{\alpha_{13}\alpha_{31}}{\Delta\epsilon^2 - \nu^2}(\cos^2 \nu t - \cos \nu t \cos \Delta\epsilon t) \right], \quad (10)
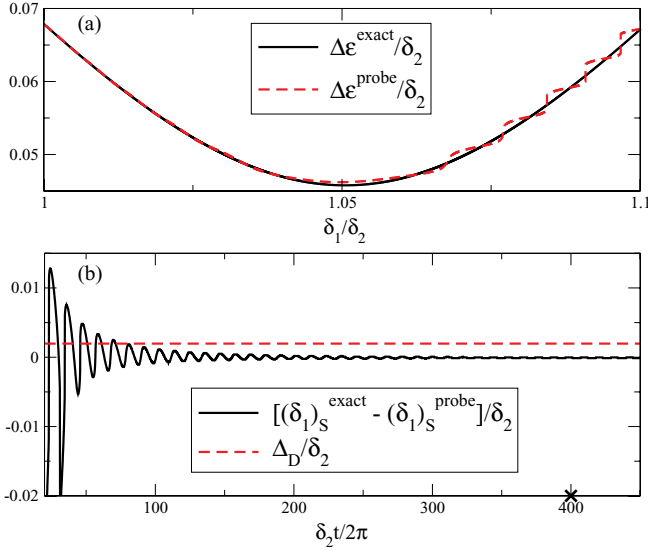\end{aligned}
$$

FIG. 4. (Color online) (a) Exact energy splitting $\Delta\epsilon = \epsilon_3 - \epsilon_2$ obtained by diagonalizing the three-level Hamiltonian (1) (black solid line), compared to the energy splitting obtained by identifying one of the maxima (in this case, the maxima around $\nu \sim \Delta\epsilon$) of the $P_{|\epsilon_2\rangle\to|\epsilon_3\rangle}$ transition probability (red dashed line). In the $t \to \infty$, limit both lines converge (the calculation is for $\delta_2 t/2\pi = 125$). (b) Difference between the exact (probeless) position of the minimum splitting (structural resonance) and the structural resonance obtained from the probed system as a function of time (black solid line). This difference goes to zero in the long-time limit as expected. The red dashed line corresponds to the exact value of the dynamical shift Δ_D, which is the precession required in the measurement of the structural resonance. The time marked by an x in (b) corresponds to $t = 2\pi/\Delta_D$, the lower bound of the time required to resolve the dynamical shift [see Eq. (11)]. $\Omega_1 = 0.2\delta_2$ and $\Omega_2 = 0.5\delta_2$.

with $\alpha_{ij} = \langle\epsilon_3|i\rangle\langle j|\epsilon_2\rangle$ and $\Delta\epsilon(\delta_1) = \epsilon_3 - \epsilon_2$. $P_{|\epsilon_2\rangle\to|\epsilon_3\rangle}$ will show peaks at $\nu \approx \pm\Delta\epsilon$. Thus, by changing the probe detuning δ_{13} (sweeping the value of ν) and measuring the corresponding transition rate for a fixed set of parameters of the probeless system, the energy splitting between levels ϵ_2 and ϵ_3 is determined. Following the same procedure for different values of δ_1, it is possible to find the minimum splitting and identify the structural resonance.

The positions of the maxima of $P_{|\epsilon_2\rangle\to|\epsilon_3\rangle}$ will only be located exactly at $\nu = \pm\Delta\epsilon$ in the long time when $\frac{\sin[(\nu\pm\Delta\epsilon)t/2]}{\nu\pm\Delta\epsilon} \xrightarrow{t\to\infty} \pi\delta(\nu\pm\Delta\epsilon)$ and the contribution of the crossed term in Eq. (10) becomes negligible. At short times, the positions of the maxima are shifted due to the δ_1 dependence of α_{ij} [see Fig. 4(a)]. To resolve the dynamical shift by this method, this effect should be smaller than the dynamical shift itself, which is indeed achieved at sufficiently long times, as shown in Fig. 4(b).

As time increases, the peaks of $P_{|\epsilon_2\rangle\to|\epsilon_3\rangle}$ become narrower (the width of each peak goes like $2\pi/t$); so, to resolve the dynamical shift, the probe beam should be applied for a time

$$t \gg \frac{2\pi}{\Delta_D}. \tag{11}$$

In summary, for large enough times, both effects (the shift due to the δ_1 dependence of α_{ij} and the width of the peaks in

order to resolve the dynamical shift) can be overcome. Note also that condition (11), which ensures narrow peaks, is more demanding than the times required to get rid of the shift due to the δ_1 dependence of the α_{ij} [see Fig. 4(b)].

The height of the peaks grows with time as $\sim \Omega_p^2 t^2/4$; to keep the perturbative treatment valid, this maximum probability has to be smaller than one (weak probe field). Combining this low-probe-intensity condition with the long-time condition given above, we end up with a condition for the probe-field amplitude Ω_p:

$$\Omega_p \ll \frac{\Omega_1^2 \Omega_2^2}{2\delta_2^3}. \tag{12}$$

Actually, this is just an upper bound. The exact growth of the height with time is given by $(\alpha_{jk})^2 \Omega_p^2 t^2/4$, but the values of the matrix elements α_{jk} are bounded between 0 and 1.

V. SPECIFIC SYSTEMS

The most obvious setting for a Raman-transition experiment is driving optical-stimulated Raman transitions in alkali-metal atoms. Unfortunately, this appears to be a difficult scenario in which to study this effect. Taking ^{87}Rb, as an example, for driving stimulated Raman transitions between hyperfine ground levels, using lasers nearly resonant with the D_2 line ($5^2S_{1/2} \to 5^2P_{3/2}$ transition), typical parameters are a detuning $\delta_1 \approx \delta_2 = 2\pi \times 10$ GHz and Rabi frequencies $\Omega_1 = \Omega_2 = 2\pi \times 200$ MHz. These parameters give a lowest-order dynamical shift [Eq. (9)] of 400 Hz. However, with an excited-state decay rate of $\Gamma = 2\pi \times 6.1$ MHz, the rate of spontaneous scattering from the Raman fields is around $R_{sc} \approx \Gamma(\Omega_1^2 + \Omega_2^2)/8\delta_1^2$, or about 3.8 kHz. The problem here is that the dressed states will be broadened at the kilohertz level, and the interaction time of the probe will be limited; therefore, the resolution of the probe will be too poor to resolve the dynamical shift. Decreasing the scattering rate also does not help much; for example, increasing the detuning to 100 GHz leads to a scattering rate of only 38 Hz, but a dynamical shift of only 400 mHz. The scattering rate becomes comparable to the dynamical shift for a detuning of only 1 GHz, which is, realistically, too small for precision measurements.

A more promising experimental realization is possible by driving microwave transitions in the hyperfine structure of the ground electronic level of atoms. Here, spontaneous emission is completely ignorable, as the magnetic-dipole transition lifetimes are much longer than any reasonable laboratory time scale. In particular, we consider here the $n^2S_{1/2}$ ground state of alkali-metal atoms, which is split into two hyperfine levels, $F = I \pm 1/2$, where I is the nuclear-spin quantum number. The three hyperfine sublevels corresponding to the setup in Fig. 1 are $|1\rangle = |F = I - 1/2, m_F = -1\rangle$ and $|3\rangle = |F = I + 1/2, m_F = -1\rangle$ for the two Raman-coupled states, and $|2\rangle = |F = I + 1/2, m_F = 0\rangle$ for the intermediate ("excited") state. The degeneracy of the $|2\rangle$ and $|3\rangle$ states is broken by applying a magnetic bias field $B_{\text{bias}} = \frac{\chi \Delta E_{\text{hfs}}}{\mu_B(g_J - g_I)}$, where ΔE_{hfs} is the zero-field hyperfine splitting, g_J and g_I are the electronic and nuclear g factors, respectively, and $\chi = (I + 1/2)^{-1}$. This represents the center of an avoided crossing of the $|1\rangle$ and $|3\rangle$ states, and thus the splitting

at this bias-field strength, $\Delta E_{31} = \sqrt{1 - \chi^2}\,\Delta E_{\rm hfs}$, is insensitive to first order to bias-field fluctuations. This reduces the need for stringent experimental control over magnetic fields, and the most important systematic error in measuring the Raman resonances. With the same magnetic field, the energy of the "excited" $|2\rangle$ state is above that of the $|3\rangle$ state by an amount given by $\Delta E_{23}(B_{\rm bias}) = g_I \mu_B B_{\rm bias} + \Delta E_{\rm hfs}(\sqrt{1 + \chi^2} - \sqrt{1 - \chi^2})/2$. For example, for ^{87}Rb, with $\Delta E_{\rm hfs} = h \times 6.835$ GHz and $I = 3/2$, the bias field is $B_{\rm bias} = 1.219$ kG, and the splittings are $\Delta E_{31} = h \times 5.919$ GHz for the (nominal) Raman resonance, and $\Delta E_{23} = h \times 860$ MHz for the Ω_2 driving transition. The remaining (Ω_1) driving transition is given by the sum of the other two transition frequencies, or $\Delta E_{21} = h \times 6.779$ GHz. Both Raman driving transitions are driven by circularly polarized fields, while the probe field is driven by a linearly polarized field.

Continuing with the ^{87}Rb example, the Raman fields may be applied with Rabi frequencies of $\Omega_1 = \Omega_2 = 2\pi \times 300$ kHz, corresponding to field intensities of about 7.6 W/cm^2 on both transitions. Microwave fields of this intensity, for example, have been realized around 6.8 GHz in the near field of an atom chip to manipulate a Bose-Einstein condensate of ^{87}Rb [3]. Thus, a field of this strength for the 6.8-GHz transition is feasible, and the field for the 860-MHz transition should similarly pose no problem. For a Raman detuning $\delta_1 \sim \delta_2 = 2\pi \times 1$ MHz, the lowest-order dynamical shift from Eq. (9) is 2.0 kHz.

In the choice of parameters here, it is also convenient to have very different Raman transition frequencies (6.8 and 0.9 GHz for the Ω_1 and Ω_2 fields, respectively) to control the secondary ac Stark shifts that we have not explicitly accounted for. For example, the 6.8-GHz Ω_1 field driving the $|1\rangle \rightarrow |2\rangle$ transition also couples the $|3\rangle \rightarrow |2\rangle$ transition at 0.9 GHz, albeit much further off resonance. As long as the δ_2 is held fixed, the Stark shift of $|1\rangle$ caused by the Ω_2 field is inconsequential, as it simply causes a common shift of both structural and dynamic resonances. However, the Stark shift of $|2\rangle$ caused by the Ω_1 field depends on δ_1, and thus can cause an additional contribution to the dynamical shift Δ_D. However, this effect is suppressed by the ratio of the detuning δ_1 from the $|1\rangle \rightarrow |2\rangle$ transition to the detuning from the $|3\rangle \rightarrow |2\rangle$ transition. This effect should thus be smaller than the lowest-order shift of 2.0 kHz by a factor of about 10^{-4}, and is therefore negligible. Note also that it is important to have Raman detunings much smaller than the transition frequencies in order to suppress the effects of Bloch-Siegert shifts. By a similar argument, the contribution of the Bloch-Siegert shifts should be of the same order as the secondary ac Stark shifts.

Uncertainties in the microwave frequencies are negligible on the scale of kilohertz for fields derived from digital synthesizers. However, the splitting at each detuning must be determined to an accuracy finer than the 2.0-kHz shift. Thus, to resolve this shift, the (6.8-GHz) probe beam should be applied for a time much longer than 500 μs [Eq. (11)], with a Rabi frequency small compared to 2π kHz [Eq. (12)]. The probe field then requires a correspondingly much lower intensity, as compared to the Raman fields. The atoms will also need to be well confined on ms time scales, without inducing spontaneous emission. Loading laser-cooled atoms into a dipole trap, formed by the focused light of a CO$_2$ laser, accomplishes this with negligible perturbation to the hyperfine structure of the ground electronic state.

Care must also be taken in preparing the atoms for the probe measurement. Since the goal is to measure the splitting of the dressed states at a particular detuning, as described above, we must prepare the atoms in only one of the dressed states. This is effected, for example, by first optically pumping the atoms (in the absence of the Raman fields, and with only a small magnetic bias field of the order of 100 mG to prevent mixing of states) into the $|1\rangle = |F = 1, m_F = -1\rangle$ bare state. This is accomplished by driving the $5^2 S_{1/2}, F = 1 \rightarrow 5^2 P_{3/2}, F' = 1$ optical transition with circularly polarized light, while optically depumping the $F = 2$ ground hyperfine level. The 1.2-kG field should then be turned on adiabatically to produce the correct level configuration without inducing any transitions. The Raman fields should then also be turned on adiabatically, but far from Raman resonance. They can then be adiabatically chirped to the desired Raman detuning, transferring the atoms from $|1\rangle$ to $|\epsilon_2\rangle$. The probe field should then be activated to attempt to drive atoms to the other dressed state $|\epsilon_3\rangle$. Finally, the Raman fields should again be detuned and adiabatically turned off, and the magnetic field turned off adiabatically as well. The population transferred to $|\epsilon_3\rangle$, and thus to $|3\rangle = |F = 2, m_F = -1\rangle$, is then measured by fluorescence detection of the $F = 2$ population. Finer resolution of the Raman splitting is also possible by employing a Ramsey-interference technique, applying the probe in two pulses separated in time.

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