Subnatural linewidths in two-photon excited state spectroscopy

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We investigate, theoretically and experimentally, absorption on an excited-state atomic transition in a thermal vapor where the lower state is coherently pumped. We find that the transition linewidth can be subnatural, that is, less than the combined linewidth of the lower and upper state. For the specific case of the 6P_1/2 → 7S_1/2 transition in room temperature cesium vapor, we measure a minimum linewidth of 6.6 MHz compared with the natural width of 8.5 MHz. Using perturbation techniques, an expression for the complex susceptibility is obtained which provides excellent agreement with the measured spectra.

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I. INTRODUCTION

Spectroscopy of excited state transitions is of growing interest for a variety of applications including the search for stable frequency references [1,2], state lifetime measurement [3], optical filtering [4], frequency up-conversion [5], multiphoton laser cooling [6], as well as Rydberg gases [7,8] and their application to electro-optics [9–11] and nonlinear optics [12]. Two-photon excited state spectroscopy can be achieved without significant transfer of population out of the ground state using electromagnetically induced transparency (EIT) [13] in the ladder configuration [14]. In conventional EIT, the excited state transition is driven by a strong coupling laser creating a transparency window which is then detected using a weak probe on the ground state transition. In thermal vapors, ladder EIT is only possible when the lower transition is probed [15] and the probe wavelength is greater than the coupling wavelength [16]. Alternatively, on strong transitions such as the infrared transitions from excited states in alkali atoms, one can probe the effects of Doppler broadening. In Sec. IV we consider a transition between two states as shown in Fig. 1(a). Neglecting the effects of Doppler broadening, it is expected that the line shape remains a Lorentzian but its FWHM is solely determined by the natural linewidth of the upper state. For the specific case of the 6P_1/2 → 7S_1/2 transition in room temperature cesium vapor, we measure a minimum linewidth of 6.6 MHz compared with the natural width of 8.5 MHz. Using perturbation techniques, an expression for the complex susceptibility is obtained which provides excellent agreement with the measured spectra.

II. EXCITED STATE TRANSITION WITH COHERENT PUMPING OF LOWER STATE

A. Equations of motion and the steady state solutions

Consider a transition between two states as shown in Fig. 1(c), a lower state |1⟩ and an upper state |2⟩ with the associated eigenenergies of hω_1 and hω_2, respectively. Initially, states |1⟩ and |2⟩ are not populated. To populate state |1⟩, the system is coherently pumped by the resonant coupling field with Rabi frequency of Ω_c from the stable eigenstate |0⟩ whose eigenenergy is hω_0 and ω_0 < ω_1 < ω_2.


\[ \mathcal{H}_0 = -\hbar \Delta_c \langle 1 | | -\hbar \Delta_R | 2 \rangle, \quad (1a) \]

with \( \Delta_p = \omega_p - (\omega_2 - \omega_1) \), \( \Delta_c = \omega_c - (\omega_1 - \omega_0) \), and \( \Delta_R = \Delta_p + \Delta_c \). Here \( \Delta_{p,c} \) is the detuning of probe (coupling) laser, \( \omega_{p,c} \) is the angular frequency of probe (coupling) laser, \( \Delta_p \) is the two-photon Raman detuning, and H.c. is the hermitian conjugate. The first term of the total Hamiltonian \( \mathcal{H}_0 \) represents the field-free atomic system, whereas the second term of the Hamiltonian \( \mathcal{H}_1 \) describes the interaction with both probe and coupling fields.

Using standard semiclassical methods [19], the equations of motion for the density matrix elements \( \rho_{ij} \) are

\[ \dot{\rho}_{00} = \Gamma_1 \rho_{11} + i \Omega_c \rho_{11} + i \Omega_p \rho_{10}, \quad (2a) \]

\[ \dot{\rho}_{11} = -\Gamma_1 \rho_{11} + \frac{i \Omega_c}{2} (\rho_{01} - \rho_{10}) + \frac{i \Omega_p}{2} (\rho_{12} - \rho_{21}), \quad (2b) \]

\[ \dot{\rho}_{22} = -\Gamma_2 \rho_{22} - \frac{i \Omega_p}{2} (\rho_{12} - \rho_{21}), \quad (2c) \]

\[ \dot{\rho}_{01} = -(i \Delta_c + \gamma') \rho_{01} - \frac{i \Omega_c}{2} (\rho_{11} - \rho_{00}) + \frac{i \Omega_p}{2} \rho_{02}, \quad (2d) \]
\[
\hat{\rho}_{12} = -(i \Delta_p + \gamma') \rho_{12} - \frac{i \Omega_p}{2} (\rho_{22} - \rho_{11}) - \frac{i \Omega_c}{2} \rho_{02}, \quad (2e)
\]
\[
\hat{\rho}_{02} = -(i \Delta_R + \gamma'') \rho_{02} + \frac{i \Omega_p}{2} \rho_{01} - \frac{i \Omega_c}{2} \rho_{12}, \quad (2f)
\]
where we define effective linewidths \(\gamma' = \Gamma_1/2 + \gamma_c\), \(\gamma'' = (\Gamma_1 + \Gamma_2)/2 + \gamma_p\), and \(\gamma''' = \Gamma_2/2 + \gamma_p + \gamma_c\), and \(\Gamma_1\) and \(\Gamma_2\) are the natural linewidths of the states \(|1\rangle\) and \(|2\rangle\), respectively. In addition to spontaneous decay, we include a dephasing due to the linewidth of the probe and coupling fields of \(\gamma_p\) and \(\gamma_c\), respectively. Solving Eqs. (2) (with \(\hat{\rho}_j = 0\)) together with the constraint \(\rho_{00} + \rho_{11} + \rho_{22} = 1\) using a perturbation technique (see Appendix A), the steady state solutions of the density matrix \(\rho_{ij}\) are given by

\[
\rho_{01} = \frac{i \Omega_c}{2} \left[ \gamma' + i \Delta_c + \frac{\Omega_c^2 \gamma' / \Gamma_1}{\gamma' - i \Delta_c} \right]^{-1}, \quad (3a)
\]
\[
\rho_{11} = \frac{\Gamma_1 \Delta_c^2 + \Gamma_1 \gamma''^2 + \gamma' \Omega_c^2}{\Gamma_1 \Delta_c^2 + \Gamma_1 \gamma''^2 + \gamma' \Omega_c^2}, \quad (3b)
\]
\[
\rho_{02} = \frac{2 \Gamma_1 (i \Delta_p + \gamma'')(i \Delta_c - \gamma') \Omega_c \rho_{00} + \gamma' \Omega_c^2 \rho_{00} + 2 \Gamma_1 \Delta_c^2 + \Gamma_1 \gamma''^2 + \gamma' \Omega_c^2) [4 (i \Delta_p + \gamma'')(i \Delta_R + \gamma''') + \Omega_c^2], \quad (3c)
\]
\[
\rho_{12} = \frac{i \Omega_c^2 \Omega_p \gamma'' / \Gamma_1 \Delta_c^2 + \Gamma_1 \gamma''^2 + \gamma' \Omega_c^2} \left[ 1 + \frac{\chi (1 + i \Delta_c / \gamma')}{\gamma'' / \gamma'''} \right] \left[ \gamma'' - i \Delta_p \right]^{-1} \left[ \gamma'' - i \Delta_p \right]. \quad (3d)
\]

In the weak excitation limit of \(\Omega_p \ll \gamma''\), \(\rho_{22} = 0\). We can therefore assume that no population is lost from the system via other decay channels from state \(|2\rangle\), although additional decay channels may contribute to the linewidth \(\Gamma_2\).

We consider \(\rho_{12}\) as it determines the complex susceptibility of the system. It is clear from Eq. (3d) that the number of atoms pumped into state \(|1\rangle\) strongly affects the magnitude of \(\rho_{12}\), manifest as the multiplication factor \(\rho_{11} \Omega_p^2 / 2\). In our case, the population of state \(|1\rangle\) is resonantly pumped from state \(|0\rangle\), that is, \(\Delta_c \approx 0\), and \(\gamma_c\) is much less than \(\gamma''\). Thus, the second term in the first square bracket approaches unity within this approximation.

**B. The complex susceptibility of the system**

The complex susceptibility of the system at the probe frequency is obtained by comparing the polarization obtained from classical electrodynamics with that calculated using a density matrix treatment [13,14]. The expression of the complex susceptibility is then given by [20]

\[
\chi = -\frac{2 N d_2^1}{\hbar \varepsilon_0 \Omega_p} \rho_{21}, \quad (4)
\]

where \(N\) is the atomic density and \(d_2^1\) is the dipole matrix element for probe transition. For \(|\chi| < 1\) the real and imaginary part of the susceptibility are respectively proportional to the refractive index \(n_R\) and the absorption coefficient \(\alpha\) via the relations

\[
n_R = 1 + \text{Re}[\chi] / 2, \quad (5a)
\]
\[
\alpha = k_p \text{Im}[\chi], \quad (5b)
\]

where \(k_p\) is the wave vector of the probe field. Thus the complex susceptibility of the system is

\[
\chi = \frac{i N \hbar d_2^1}{\hbar \varepsilon_0} \left[ \gamma'' - i \Delta_R + \frac{\Omega_c^2 / 4}{\gamma'' - i \Delta_p} \right]^{-1}. \quad (6)
\]

Here \(N_1 = N \rho_{11}\), the atomic density of state \(|1\rangle\). According to Eq. (6), the complex susceptibility is proportional to the number of the atoms in state \(|1\rangle\). As state \(|1\rangle\) is coherently pumped from state \(|0\rangle\), the multiplications factor has a Lorentzian profile as a function of \(\Delta_c\). This implies that to get a large susceptibility, one needs to resonantly pump population

![Figure 1](image-url)
to state $|1\rangle$. The term in the square bracket is similar to the result obtained for EIT in the three-level cascade system [14]. It indicates that, for a finite value of $\Omega_c$, the system will become transparent when the probe field is scanned across the resonance at $\Delta_p = 0$ due to Autler-Townes splitting [21]. When state $|1\rangle$ is weakly pumped by the coupling field, that is, $\Omega_c^2 \ll \Gamma_2 (\Gamma_1 + \Gamma)$, the term in the square bracket of Eq. (6) can be expanded using a Taylor expansion. Neglecting the higher-order terms in $\Omega_c$, the complex susceptibility reduces to

$$
\chi = \frac{i N d^{2}_{21}}{\hbar \epsilon_0} \left[ \frac{1}{\gamma'' - i \Delta_R} \right].
$$

This complex susceptibility is similar to that of a two-level system, except for the multiplication factor. The transmission line shape of the system is simply a Lorentzian centered at $-\Delta_p$ with FWHM $\gamma''$. For a sufficiently small laser linewidth compared to $\Gamma_1$, the approximation of the FWHM is solely determined by the linewidth of the excited state $\Gamma_2$ irrespective of the linewidth of state $|1\rangle \Gamma_1$, that is,

$$
\Gamma_{\text{FWHM}} = \Gamma_2.
$$

This result is different to the case in which state $|1\rangle$ is incoherently populated. In such a case, the FWHM of the transmission line shape is determined by the sum of the linewidths from both the lower state and the upper state, that is, $\Gamma_1 + \Gamma_2$ [22].

For experiments in room temperature vapors, the Doppler effect must be included into the model and this topic will be discussed in the next section.

III. EFFECT OF DOPPLER BROADENING

Each atomic velocity class in a thermal vapor experiences a different laser detuning $\Delta_p$ and $\Delta_c$ due to the Doppler effect. To obtain the velocity-dependent complex susceptibility, we make changes to $\Delta_p$, $\Delta_c$, and $N$ with the following substitutions [14]:

$$
\begin{align*}
\Delta_p &\rightarrow \Delta_p - k_p v, \\
\Delta_c &\rightarrow \Delta_c + k_c v, \\
N &\rightarrow \frac{N}{u \sqrt{\pi} \exp \left( - \frac{u^2}{\mu^2} \right)}.
\end{align*}
$$

where $k_{\text{pc}}$ is the wave vector of the probe (coupling) field, $u = \sqrt{2 m k_B T / m}$ is the most probable speed of the atoms at a given temperature $T$ and $m$ is the mass of an atom. Substituting Eqs. (9) into Eq. (6), the value of the complex susceptibility of a particular velocity class $v$ is then given by

$$
\chi(v) dv = -\frac{N d^{2}_{21} \Omega_c^2}{\hbar \epsilon_0 \sqrt{\pi} k_c^2 (k_c - k_p) u^3} \Gamma_1 \left[ \frac{e^{-z^2}}{(z + \beta)^2 + \sigma^2} \right] \left[ z - z_0 + \frac{\Omega_c^2/4}{(k_c - k_p) k_p u^2 (z - z_1)} \right]^{-1} dz,
$$

with the change of variable $z = v / u$, and

$$
\begin{align*}
\gamma' &= \frac{\gamma'' (k_c - k_p) u}{(k_c - k_p) u}, \\
\sigma &= \frac{1}{k_c} u \sqrt{\gamma^2 + \frac{\Omega_c^2 \gamma'}{\Gamma_1}}, \\
\xi &= \frac{\Delta_R}{(k_c - k_p) u}, \\
\beta &= \frac{\Delta_c}{k_c u}, \\
z_0 &= -\xi - i \gamma', \\
z_1 &= \frac{\Delta_p + i \gamma''}{k_p u}.
\end{align*}
$$

The total susceptibility is obtained by integrating Eq. (10) over all velocity classes. The full result of the integration is discussed in Appendix B. We consider the case in which the coupling Rabi frequency $\Omega_c$ is sufficiently weak that the EIT-like third term in the square bracket of Eq. (10) is neglected. In this case the complex susceptibility $\chi_D$ becomes

$$
\chi_D(\Delta_p) = -\frac{N d^{2}_{21} \Omega_c^2}{\hbar \epsilon_0 \sqrt{\pi} k_c^2 (k_c - k_p) u^3} \gamma' \int_{-\infty}^{\infty} \frac{e^{-z^2}}{(z + \beta)^2 + \sigma^2} dz.
$$

From Eq. (12), the total complex susceptibility is simply the convolution between a Lorentzian of width $\gamma$ (term in square bracket describing the transition from lower state $|1\rangle$ to upper state $|2\rangle$) and a product of a Lorentzian of width $\sigma$ and a Gaussian (term in curly bracket describing the atomic velocity distribution of lower state $|1\rangle$).

The result of the integration in Eq. (12) involves the Faddeva function [23] (the exact result of the integration is described in Appendix C). However, the integration can be simplified by replacing the product between a Gaussian and a Lorentzian function [23] (the exact result of the integration is described in Appendix C). However, the integration can be simplified by replacing the product between a Gaussian and a Lorentzian by a convolution of a Gaussian and a Lorentzian with the FWHM $\gamma''$. This approximation is valid since at room temperature the Doppler effect is weak and we can approximate the product by the Lorentzian of width $\sigma$. In other words, the range of velocity classes involved in the integration around the position where the Lorentzian of width $\sigma$ is centered is much smaller than the most probable speed of the atoms $v \ll u$. Hence we can expand the Gaussian around the position where the Lorentzian is centered, that is, $\exp(-z^2) \approx \exp(-\beta^2 z)$. Using this approximation, the total susceptibility is simply given by

$$
\chi_D(\Delta_p) = -\frac{N d^{2}_{21} \Omega_c^2 \sqrt{\pi}}{\hbar \epsilon_0 k_c^2 u \sigma} \frac{1}{2 \Gamma_1 \Delta_R + i \Gamma_{\text{FWHM}} / 2},
$$

for experiments in room temperature vapors, the Doppler effect must be included into the model and this topic will be discussed in the next section.

III. EFFECT OF DOPPLER BROADENING

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with
\[
\frac{\Gamma_{\text{FWHM}}}{2} = \gamma'' + \left(\frac{k_e - k_p}{k_c}\right) \sqrt{\gamma'^2 + \frac{\Omega_2^2 \gamma'}{\Gamma_1}}. \tag{15}
\]

It is clear from Eqs. (14) and (15) that the absorption line shape remains Lorentzian with the FWHM of \(\Gamma_{\text{FWHM}}\) even when the Doppler effect is included. It is worth noting that the total susceptibility in this case is different from the total susceptibility calculated for the case of incoherent pumping. The total susceptibility for incoherent pumping is the convolution between a Lorentzian and a Gaussian, resulting in a Voigt profile [24].

In the limit where \(\Omega_c/\Gamma_1 \ll 1\), the linewidth of the absorption profile is simply
\[
\Gamma_{\text{FWHM}} = \Gamma_2 + \left(\frac{k_e - k_p}{k_c}\right) \Gamma_1 \tag{16}
\]
(neglecting \(\gamma_p\) and \(\gamma_c\)). It contains the sum of two terms: the first term is the linewidth of the absorption line shape in the case in which the Doppler effect is neglected and the latter is the linewidth of the lower state scaled by the ratio obtained from the wave vectors. Physically the second term originates from the fact that the atoms are velocity selected by the Doppler effect for the atom-field interaction. Only atoms whose velocities are between \(-\Gamma_1/2k_c\) and \(\Gamma_1/2k_c\) are coherently pumped into the lower state when the coupling field is on resonance. Since \(\Gamma_1/k_c\) is very small compared to the width of the Doppler broadening, all atoms pumped into state \([1]\) have approximately the same velocity distribution, that is, the distribution is independent of velocity, and given by
\[
f(v) = \frac{1}{\hbar \sqrt{\pi}} e^{-\beta^2}. \tag{17}
\]
However, the distribution of the atoms is also determined by the Lorentzian of width \(\Gamma_1/k_c\). Thus the final velocity distribution of the pumped atoms is a Lorentzian of width \(\Gamma_1/k_c\) and the height is scaled by Eq. (17). In the two-photon interaction process, the width of \(\Gamma_1/k_c\) in velocity space is equivalent to the width of \((k_e - k_p)\Gamma_1/k_c\) in frequency space. Hence the total linewidth of the final absorption line shape is the sum of \((k_e - k_p)\Gamma_1/k_c\) with the unaffected linewidth \(\Gamma_2\).

IV. COMPARISON BETWEEN THEORY AND EXPERIMENTAL RESULTS

To test the result derived above we used the experimental setup described by Carr et al. [25]. The experiment was performed in a 7.5 cm vapor cell containing Cs at room temperature. The 1470 nm weak probe beam (with horizontal linear polarization) and the 852 nm coaxial, counterpropagating coupling beam (with horizontal linear polarization) are applied along the vapor cell axis. The probe and coupling beams have \(1/e^2\) radii of 1.2 and 1.6 mm, respectively. The coupling beam was stabilized to the \(6S_{1/2}, F = 4 \rightarrow 6P_{3/2}, F' = 5\) transition while the probe beam was scanned across the \(6P_{3/2}, F'' = 5 \rightarrow 7S_{1/2}, F'' = 4\) transition. The scan is calibrated using a wavemeter to better than 1% accuracy. The transmission signal measured from the experiment is shown as the solid black line in Fig. 2(a).

FIG. 2. (Color online) (a) Comparison between the observed spectra, shown by a black solid line, for a relatively small \(\Omega_c' = 0.6\) MHz, and the theoretical model. The gray (blue) solid line is the theoretical prediction, taking into account the absorption of the coupling field across the vapor cell. (b) The residual plot between the observed data and the theoretical model.

To model the transmission line shape, the complex susceptibility is calculated for each magnetic sublevel and the total complex susceptibility is the average of all complex susceptibilities over all possible magnetic sublevels. This is given by
\[
\chi_{\text{tot}}(\Delta_p) = \frac{1}{16} \sum_{m_F=-4}^{4} \chi_D^{m_F}(16), \tag{18}
\]
where \(\chi_D^{m_F}(\Delta_p)\) is the complex susceptibility corresponding to the \(m_F\) magnetic sublevel of \(6S_{1/2}, F = 4\) state. \(\chi_D^{m_F}\) is calculated using Eq. (14), where the coupling Rabi frequency is sufficiently weak. The factor of \(1/16\) in the equation accounts for the fact that the initial population is evenly distributed among the magnetic sublevels of \(6S_{1/2}, F = 3, 4\). The coupling Rabi frequency of the transition \(6S_{1/2}, F = 4 \rightarrow 6P_{3/2}, F' = 5\) and the dipole matrix element of the transition \(6P_{3/2}, F'' = 5 \rightarrow 7S_{1/2}, F'' = 4\) corresponding to each magnetic sublevel \(\Omega_c^{m_F}\) and \(d_{21}^{m_F}\) are given by
\[
\Omega_c^{m_F} = \Omega_c' \times \sqrt{\Pi} \begin{pmatrix} 5 & 4 & 4 \\ m_F & 0 & -m_F \end{pmatrix}, \tag{19a}
\]
\[
d_{21}^{m_F} = 5.63 e a_0 \times \sqrt{\Pi} T \begin{pmatrix} 5 & 4 & 4 \\ m_F & 0 & -m_F \end{pmatrix}, \tag{19b}
\]
where the reduced dipole matrix element of the transition \(6S_{1/2}, F = 4 \rightarrow 6P_{3/2}, F' = 5\) is absorbed into \(\Omega_c'\), that is, \(\Omega_c' = e E_z (6P_{3/2})r || 6S_{1/2}/r, a_0\) is Bohr radius, and \(m_F\) and \(m_F\) are the magnetic sublevels of the \(6S_{1/2}\) and \(6P_{3/2}\), respectively.

The comparison between the experimental data and the theoretical model is shown in Fig. 2(a). The theoretical transmission shown as the solid gray (blue) line was calculated using Eqs. (19b) and (18) for a known temperature of \(T = 22\) °C. The fit parameters \(\Omega_c'/(2\pi) = 0.6\) MHz, \(\gamma_p/(2\pi) = 0.2\) MHz are
FIG. 3. (Color online) The values of the FWHM are plotted as a function of $\Omega_c/2\pi$. The dashed curve is the theoretical prediction calculated using the weak coupling model, whereas the solid (red) curve is the theoretical prediction calculated using the full model. As expected the data are in agreement with the weak coupling model when $\Omega_c/2\pi$ is relatively small. However, for a large $\Omega_c/2\pi$, the weak coupling model fails to predict the FWHM. This difference at large $\Omega_c/2\pi$ can be recovered when using the full model. The combined lower and upper state linewidth $\Gamma_1 + \Gamma_2$ is shown by the dotted line.

FIG. 4. (Color online) (a) Comparison between the observed spectra, shown by a black solid line, for $\Delta p/2\pi = 3.0$ MHz, and the theoretical model. The gray (blue) dotted line is the theoretical prediction calculated using the weak coupling model, whereas the gray (red) solid line is the theoretical prediction calculated using the full model. (b) The residual plot between the observed data and the theoretical model calculated using the full model.

FIG. 5. (Color online) (a) Comparison between the observed spectra, shown by a black solid line, for $\Delta p/2\pi = 15$ MHz, and the theoretical model. The gray (red) solid line is the theoretical prediction calculated using the full model. (b) The residual plot between the observed data and the theoretical model.
to Autler-Townes split the absorption resonance. The value of $\Omega_p^*/2\pi$ in this case is 15 MHz. The gray (red) solid line is the theoretical prediction calculated using the full model in the region $|\Delta_p/2\pi| \ll 20$ MHz. Both the theoretical prediction and the observed data are in good agreement around resonance. Figure 5(b) shows the residual between the theoretical prediction and the observed data.

VI. CONCLUSION

We have developed the theory of absorption line shapes on excited state transitions where the lower state is coherently populated. We show that for an atom at rest, in the limit of weak pumping, the line shape is a Lorentzian and the linewidth of the transition reduces to the linewidth of the upper state. Including the effect of Doppler broadening the line width is still subnatural and we find that the predicted line shape is in very good agreement with experimental data over a wide range of coupling field parameters.

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APPENDIX A: STEADY STATE SOLUTIONS BY PERTURBATION TECHNIQUE

Since the probe field is sufficiently weak (it will be shown later that the weak probe condition is fulfilled when $\Omega_p/\gamma' \ll 1$), one can consider the expansion of the density matrix $\rho_{ij}$ in the power of $\Omega_p$, namely,

$$\rho_{ij} = \rho_{ij}^{(0)} + \rho_{ij}^{(1)} \Omega_p + \rho_{ij}^{(2)} \Omega_p^2 + \rho_{ij}^{(3)} \Omega_p^3 + \cdots, \quad (A1)$$

where $\rho_{ij}^{(n)}$ is the $n$th order correction of the expansion of $\rho_{ij}$. To solve for $\rho_{ij}$, we substitute Eq. (A1) into Eqs. (2), equate the terms of the same power in $\Omega_p$, and then solve for $\rho_{ij}^{(n)}$ from $n = 0$ to all $n$ [27].

Applying this technique to Eqs. (2), the set of equations corresponding to the zeroth power of $\Omega_p$ is given by

$$\Gamma_1 \rho_{11}^{(0)} + \frac{i}{2} \Omega_{p} (\rho_{01}^{(0)} - \rho_{10}^{(0)}) = 0, \quad (A2a)$$

$$\Gamma_2 \rho_{22}^{(0)} = 0, \quad (A2b)$$

$$(i \Delta_c + \gamma') \rho_{01}^{(0)} + \frac{i}{2} \Omega_c (\rho_{11}^{(0)} - \rho_{00}^{(0)}) = 0, \quad (A2c)$$

$$(i \Delta_p + \gamma') \rho_{12}^{(0)} + \frac{i}{2} \Omega_c \rho_{02}^{(0)} = 0, \quad (A2d)$$

$$(i \Delta_R + \gamma'') \rho_{02}^{(0)} + i \frac{2}{\Omega_c} \rho_{02}^{(0)} = 0, \quad (A2e)$$

$$\rho_{00}^{(0)} + \rho_{11}^{(0)} + \rho_{22}^{(0)} = 1. \quad (A2f)$$

We find that the zeroth order corrections of $\rho_{ij}$ vanish, except $\rho_{00}^{(0)}$, $\rho_{11}^{(0)}$, and $\rho_{02}^{(0)}$. The expressions for $\rho_{00}^{(0)}$ and $\rho_{11}^{(0)}$ are given by

$$\rho_{00}^{(0)} = \frac{i \Omega_c}{2} \left[ \frac{\Delta_p - \gamma' \rho_{11}^{(0)}}{\gamma' - i \Delta_c} \right], \quad (A3a)$$

$$\rho_{11}^{(0)} = \frac{\Omega_c^2 \gamma'/\Gamma_1}{\Gamma_1 \Delta_c^2 + \Gamma_1 \gamma'^2 + \gamma' \Omega_c^2}. \quad (A3b)$$

Similarly, the set of equations corresponding to the nth power of $\Omega_p$ (for $n \geq 1$) is given by

$$\Gamma_1 \rho_{11}^{(n)} + \frac{i}{2} \Omega_c (\rho_{01}^{(n)} - \rho_{10}^{(n)}) = 0, \quad (A4a)$$

$$\Gamma_2 \rho_{22}^{(n)} + \frac{i}{2} \rho_{12}^{(n-1)} - \rho_{21}^{(n-1)} = 0, \quad (A4b)$$

$$(i \Delta_c + \gamma') \rho_{01}^{(n)} - \frac{i}{2} \rho_{02}^{(n-1)} + \frac{i}{2} \Omega_c (\rho_{11}^{(n)} - \rho_{00}^{(n)}) = 0, \quad (A4c)$$

$$(i \Delta_p + \gamma'') \rho_{02}^{(n)} + i \frac{2}{\Omega_c} \rho_{02}^{(n)} + i \frac{2}{\Omega_c} (\rho_{12}^{(n-1)} - \rho_{11}^{(n-1)}) = 0, \quad (A4d)$$

$$(i \Delta_R + \gamma'') \rho_{02}^{(n)} - \frac{i}{2} \rho_{01}^{(n-1)} + i \frac{2}{\Omega_c} \rho_{12}^{(n-1)} = 0, \quad (A4e)$$

$$\rho_{00}^{(n)} + \rho_{11}^{(n)} + \rho_{22}^{(n)} = 0. \quad (A4f)$$

Using Eqs. (A3) and (A4), all of $\rho_{ij}^{(1)}$ again vanish, except $\rho_{02}^{(1)}$ and $\rho_{12}^{(1)}$, whose expressions are given by

$$\rho_{02}^{(1)} = \frac{2 \Gamma_1 (i \Delta_p + \gamma')(i \Delta_c - \gamma') \Omega_c + \gamma' \Omega_c^3}{2 \Gamma_1 \Delta_c^2 + \Gamma_1 \gamma'^2 + \gamma' \Omega_c^2} \left[ 4(i \Delta_p + \gamma')(i \Delta_R + \gamma'') + \Omega_c^2 \right]. \quad (A5a)$$

$$\rho_{12}^{(1)} = \frac{i \Omega_c^2 \gamma'/4}{\Gamma_1 \Delta_c^2 + \Gamma_1 \gamma'^2 + \gamma' \Omega_c^2} \left[ 1 + \frac{\gamma_c (1 + i \Delta_c / \gamma')}{\gamma'' + i \Delta_p} \right] \left[ \gamma'' + i \Delta_R + \frac{\Omega_c^2 / 4}{\gamma'' + i \Delta_p} \right]. \quad (A5b)$$

It can be shown that $\rho_{12}^{(2)} = \rho_{21}^{(2)} = 0$. The solutions of the coherence $\rho_{02}$ and $\rho_{12}$, to second order, are then

$$\rho_{02} = \Omega_p \rho_{02}^{(1)}, \quad (A6a)$$

$$\rho_{12} = \Omega_p \rho_{12}^{(1)}. \quad (A6b)$$

Thus far we have assumed only that the probe Rabi frequency is sufficiently weak without quantifying this condition. To quantitatively determine the weak probe condition, let us consider the steady state of $\rho_{00}$ as this quantity is strongly related to $\rho_{11}$ and hence also to $\rho_{12}$. Using Eq. (2d) with the substitution $\rho_{00} = 1 - \rho_{11} - \rho_{22}$, the expression of the steady
state of $\rho_{01}$ given by
\[
\rho_{01} = -\frac{i\Omega_c\rho_{11}}{i\Delta_c + \gamma'} - \frac{i\Omega_c\rho_{22}/2}{i\Delta_c + \gamma'} + \frac{i\Omega_c/2}{i\Delta_c + \gamma'} + \frac{i\Omega_p\rho_{02}/2}{i\Delta_c + \gamma'}.
\tag{A7}
\]

The first approximation is to neglect the contribution to $\rho_{01}$ from $\rho_{22}$ as $\rho_{22}$ vanishes up to the second order correction. This approximation is justified as the probe laser is weak and the upper state population is negligible. Thereafter we consider the last term which contains the product between $(\Omega_p/2)/(i\Delta_c + \gamma')$ and $\rho_{02}$. The first term is of order $\Omega_p/\gamma'$ at resonance $\Delta_c = 0$. The second term $\rho_{02}$ is also of order $\Omega_p/\gamma'$ at resonance [see Eqs. (A5a) and (7a)]. Thus the product of these two terms is of order $(\Omega_p/\Gamma)^2$, which can be neglected if $\Omega_p/\gamma'$ is much less than one. It follows that these approximations lead to the same results for $\rho_j$ given by Eqs. (8). Hence, the weak probe condition is valid when $\Omega_p/\gamma' \ll 1$.

**APPENDIX B: COMPLETE SOLUTION OF THE COMPLEX SUSCEPTIBILITY OF THE SYSTEM**

The complete solution of the complex susceptibility can be found by evaluating the integral
\[
\int_{-\infty}^{\infty} \frac{e^{-z^2}}{(z + \beta)^2 + \sigma^2} \left[ z - z_0 + \frac{\Omega_p^2/4}{(k_c - k_p)k_p u^2(z - z_1)} \right] dz.
\tag{B1}
\]

To evaluate the integral, one rewrites the integrand, using partial fractions as
\[
\frac{e^{-z^2}}{(z + \beta)^2 + \sigma^2} \left[ z - z_0 + \frac{\Omega_p^2/4}{(k_c - k_p)k_p u^2(z - z_1)} \right]^{-1} = -\frac{(z_1 - \phi_+)}{[(\beta + \phi_+)^2 + \sigma^2](\phi_+ - \phi_-) z - \phi_+}
+ \frac{(z_1 - \phi_-)}{[(\beta + \phi_-)^2 + \sigma^2](\phi_+ - \phi_-) z - \phi_-}
+ \frac{i(z_1 + \beta + i\sigma)}{2\sigma(\beta + \phi_+ + i\sigma)(\beta + \phi_+ - i\sigma) z - \beta + i\sigma}
+ \frac{i(z_1 + \beta - i\sigma)}{2\sigma(\beta + \phi_- - i\sigma)(\beta + \phi_- + i\sigma) z - \beta - i\sigma}.
\tag{B2}
\]

where
\[
\phi_{\pm} = \frac{1}{2}(z_0 + z_1) \pm \frac{1}{2\sqrt{(z_0 - z_1)^2 - \Omega_p^2/k_p(k_c - k_p)u^2}}.
\tag{B3}
\]

The integrand has four poles in the complex plane, that is, at $\phi_+, \phi_- - \beta - i\sigma$, and $-\beta + i\sigma$. Clearly this complex integral reduces to the integral of the form
\[
\int_{-\infty}^{\infty} \frac{e^{-z^2}}{z - z_p} dz.
\tag{B4}
\]

where $z_p$ is the pole in the complex plane. The solution of the integration is given by
\[
\int_{-\infty}^{\infty} \frac{e^{-z^2}}{z - z_p} dz = is\pi W(sz_p),
\tag{B5}
\]

where $s = \text{sgn}[\text{Im}(z_p)]$ and $\text{sgn}$ is known as the signum function and its value is $+1$ when the argument is positive and $-1$ when the argument is negative. $W(z)$ is known as the Faddeva function and it is defined as
\[
W(z) = e^{-z^2} \text{erfc}(-iz).
\tag{B6}
\]

Hence the complex susceptibility is given by
\[
\chi_D = -\frac{iNd^2\Omega_p^2\sqrt{\pi}}{h\epsilon_0k_0^2(k_c - k_p)u^3} \times \left[ -\frac{(z_1 - \phi_+)}{\pi(\beta + \phi_+)^2 + \sigma^2} s_+ W(s_+\phi_+) + \frac{(z_1 - \phi_-)}{\pi(\beta + \phi_-)^2 + \sigma^2} s_- W(s_-\phi_-) - \frac{2\sigma(\beta + \phi_+ + i\sigma)(\beta - \phi_- + i\sigma)}{\pi\sigma(\phi_+ - \phi_-)(\beta + \phi_+ - i\sigma)} W(-\beta - i\sigma) + \frac{2\sigma(\beta + \phi_- - i\sigma)(\beta + \phi_- + i\sigma)}{\pi\sigma(\phi_+ - \phi_-)(\beta - \phi_- + i\sigma)} W(-\beta + i\sigma) \right].
\tag{B7}
\]

where $s_+ = \text{sgn}[\text{Im}(\phi_+)]$ and $s_- = \text{sgn}[\text{Im}(\phi_-)]$.

One can use the same approximation as discussed in the article to approximate Eq. (B7) and the approximated expression of Eq. (B7) is given by
\[
\chi_D = -\frac{Nd^2\Omega_p^2}{h\epsilon_0\sqrt{\pi}k_0^2(k_c - k_p)u^3} e^{-\beta^2} \times \left[ \frac{\pi(\beta + z_1)(\phi_+ - \phi_-) + is_+\pi\sigma(\phi_+ + \phi_- - 2z_1)}{\pi(\phi_+ - \phi_-)(\beta + \phi_+ - i\sigma)} \right].
\tag{B8}
\]

**APPENDIX C: THE EXACT RESULT OF EQ. (13)**

To evaluate the integral in Eq. (13), the integrand can be rewritten using partial fractions as
\[
\frac{e^{-z^2}}{(z + \beta)^2 + \sigma^2} = -\frac{e^{-z^2}/2}{\sigma(\beta - \gamma + i\xi)(\beta - i\sigma)} e^{-z^2} + \frac{e^{-z^2}/2}{\sigma(\beta + \gamma - i\xi)(\beta + i\sigma)} e^{-z^2} + \frac{\sigma(\beta + \gamma + i\xi)(\beta + i\sigma)}{[\sigma^2 + (\xi + i\gamma)^2]}(z + \xi + i\gamma).
\tag{C1}
\]

Using Eq. (B5) and rearranging the expression, the complex susceptibility is given by
\[
\chi_D(\Delta_p) = -\frac{iNd^2\Omega_p^2\sqrt{\pi}}{4h\epsilon_0k_0^2(k_c - k_p)u^3} \times \left[ e^{-z_p^2} \text{erfc}(-iz_0) + \frac{i\xi_0}{\sigma} e^{z_p^2} \text{erfc}(\sigma) \right],
\tag{C2}
\]

where $z_0 = \xi + i\gamma$. 033830-7