Effects of Doppler broadening on Autler-Townes splitting in six-wave mixing

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The effects of Doppler broadening on Autler-Townes (AT) splitting in six-wave mixing (SWM) are investigated by the dressed-state model. We analyze the velocities at which the atoms are in resonance with the dressed states through Doppler frequency shifting and find that, depending on the wave-number ratio, there may be two resonant velocities which can originate from resonance with one of the dressed states or from resonance with two different dressed states. Based on this model, we discuss a novel type of AT doublet in the SWM spectrum, where macroscopic effects play an important role. Specifically, the existence of resonant peaks requires polarization interference between atoms of different velocities in addition to a change in the number of resonant atoms involved. Our model can also be employed to analyze electromagnetically induced transparency resonance and other types of Doppler-free high-resolution AT spectroscopy.

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I. INTRODUCTION

When a two-level system is driven by a strong coupling field at a resonant frequency, the populations of the states undergo coherent Rabi oscillations. This coherent process is reflected in the appearance of two sidebands offset by the Rabi frequency from the main transition. The phenomenon can be described in terms of dressed states, which are eigenstates of the total system of an atom plus the coupling field. Dressed states are usually probed through a transition to or from a third level as a doublet excitation spectrum, called an Autler-Townes (AT) doublet [1]. The AT doublet has been demonstrated in both atomic [2] and molecular systems [3–5]. More recently, they have also been observed in quantum dots [6–8] and in superconducting qubits [9,10].

One important application of the AT effect is that it can be used to measure directly the absolute value of the molecular transition dipole moment [3] and its dependence on the internuclear distance [11]. On the other hand, the effects of inhomogeneous Doppler broadening on the AT splitting have been investigated. Ahmed and Lyyra [12] analyzed theoretically the AT splitting in a Doppler-broadened three-level cascade system. They found that the observed AT splitting is not only a function of the coupling-laser Rabi frequency, as in the homogeneously broadened case, but can also strongly depend on the wave-number ratio of the coupling and probe lasers. Relevant experiments have been performed in a three-level Na_2 open molecular cascade system where, for moderate Rabi frequencies, the fluorescence line shape from the uppermost level in the system depends strongly on the wave-number ratio of the two laser fields [13].

Traditionally, an AT doublet was observed through probe absorption or fluorescence excitation spectroscopy. Recently, Doppler-free four-wave mixing spectroscopy has been employed for probing dressed states [14–16]. Moreover, higher-order wave mixing has attracted much attention [17–20]. In our previous works, we studied the AT splitting in electromagnetically-induced-transparency-based (EIT-based) six-wave mixing (SWM) in a Doppler-broadened system [21,22]. An AT doublet appears in the SWM spectrum when atoms with specific velocities have double resonance with one of the dressed states and with a third level.

In this paper we study a novel type of AT splitting, where atoms with certain velocities are only singly resonant with one of the dressed states. For an AT doublet to appear in the SWM spectrum, there must be some macroscopic effects involved. Specifically, atoms in a wide region of velocities can contribute to the SWM signal at resonance, whereas when the incident laser frequency is detuned away from resonance, destructive interference between the polarizations of atoms of different velocities causes strong suppression of the SWM signal. We have also employed the dressed-state model to explain the strong wave-number-ratio dependence of the AT splitting. For example, in the presence of a strong coupling field the SWM spectrum exhibits either a single peak or an AT doublet, depending on the ratio between the magnitudes of the wave vectors [22]. In this paper, we analyze the velocities at which atoms are in resonance with the dressed states through Doppler frequency shifting. It is found that, depending on the wave-number ratio, the two resonant velocities can originate either from the resonance with one of the dressed states or from that with two different dressed states. On the other hand, in some cases there will be no atoms which can resonate with the dressed states. Based on these features, the strong dependence of the SWM spectra on the wave-number ratio can be explained. Finally, we point out that our model can also be employed to explain the EIT resonance in a Doppler-broadened three-level system and other types of Doppler-free high-resolution AT spectroscopy.

Generally speaking, this work is a continuation of the studies of Doppler effects in media with excited coherence. For example, the effect of Doppler broadening on the width of an EIT resonance has been investigated by Javan and coworkers [23,24], and that on the group velocity in a slow-light medium has also been studied [25,26]. In particular, Scully and coworkers [27,28] have demonstrated three-photon electromagnetically induced absorption and transparency in rubidium atomic vapor driven by two coherent electromagnetic fields. They observed narrow absorption as well as a transmission window on the background of high-contrast Doppler-free subnatural absorption resonance. The Doppler-free resonances originate from the behavior of dressed states in coherent fields. On the other hand, the effects of Doppler broadening on a generalized double-dark resonance have been investigated by Ye *et al.* [29].

This paper is organized as follows: Section II presents the basic theory of EIT-based SWM in Doppler-broadened four-level systems. The expression for the nonlinear polarization responsible for the SWM signal is derived, based on which conditions for observing AT splitting in the SWM spectrum of a Doppler-broadened system are analyzed. We then use a dressed-state model in Sec. III to analyze the velocities at which atoms are in resonance with dressed states through Doppler frequency shifting. The wave-number-ratio dependence of the AT splitting is explained with this model. In Sec. VI, we focus on the EIT-based SWM in a Doppler-broadened folded four-level system. The anomalous resonance, which involves macroscopic effects, is analyzed in detail. Finally, Sec. V is the discussion and conclusion. We point out that our model can be employed to explain the EIT resonance in a Doppler-broadened three-level system and for other types of Doppler-free high-resolution AT spectroscopy.

II. EIT-BASED SWM IN DOPPLER-BROADENED FOUR-LEVEL SYSTEMS

Let us consider a cascade system and a folded four-level system (Fig. 1), where the states between $|0\rangle$ and $|1\rangle$, $|1\rangle$ and $|2\rangle$, and $|2\rangle$ and $|3\rangle$ are coupled by dipolar transitions with resonant frequencies Ω_1, Ω_2, and Ω_3 and dipole moment matrix elements μ_1, μ_2, and μ_3, respectively. In SWM, beams 3 and 3′ have the same frequency ω_3 and a small angle exists between them. Beam 1 with frequency ω_1 propagates along the opposite direction to beam 3, while beam 2 with frequency ω_2 can propagate either along or in the opposite direction. We assume that $\omega_i \simeq \Omega_i$ so that ω_i drives the transition from $|i-1\rangle$ to $|i\rangle$. The simultaneous interactions of atoms with beams 1, 2 and 3 will induce atomic coherence between $|0\rangle$ and $|3\rangle$ through a resonant three-photon transition. This three-photon coherence is then probed by beams 3′ and 2, and as a result a SWM signal of frequency ω_1 is generated almost opposite to the direction of beam 3′.

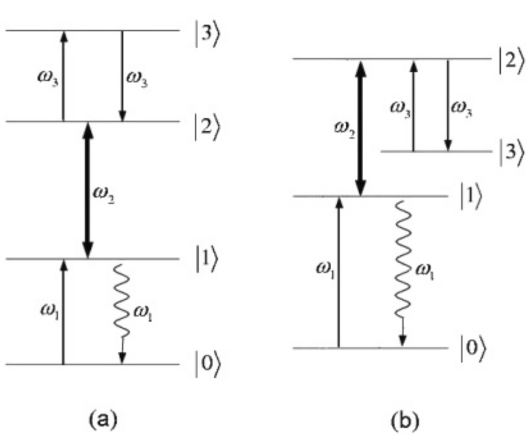

FIG. 1. Energy-level diagram for resonant SWM in (a) cascade and (b) folded four-level systems.

Let the detunings be represented by $\Delta_i = \Omega_i - \omega_i$, so after a canonical transformation we have

$$\begin{aligned} H^{(\pm)} &= \hbar\Delta_1|1\rangle\langle 1| + \hbar(\Delta_1+\Delta_2)|2\rangle\langle 2| \\ &+\hbar(\Delta_1+\Delta_2\pm\Delta_3)|3\rangle\langle 3| \\ &-[\mu_1 E_1|1\rangle\langle 0| + \mu_2 E_2|2\rangle\langle 1| \\ &+\mu_3(E_3+E_3')|3\rangle\langle 2| + \text{H.c.}]. \end{aligned} \tag{1}$$

Here, $H^{(+)}$ and $H^{(-)}$ are the effective Hamiltonians for the cascade and folded four-level systems, respectively; the quantities $E_i = \varepsilon_i e^{i\mathbf{k}_i\cdot\mathbf{r}}$ ($i = 1$ to 3) and $E_3' = \varepsilon_3' e^{i\mathbf{k}_3'\cdot\mathbf{r}}$ are the complex incident laser fields, where $\mathbf{k}_i$ and $\mathbf{k}_3'$ are the wave vectors of beams i and 3′, respectively. In a Doppler-broadened system, the nonlinear polarization responsible for the SWM signal is given by

$$P^{(\pm)} = iN\mu_1 G_1|G_2|^2 G_3(G_3')^* \int_{-\infty}^{\infty} d\mathbf{v}W(\mathbf{v})F^{(\pm)}(\mathbf{v}). \tag{2}$$

Here, $W(\mathbf{v}) = [1/(\sqrt{\pi}u)]e^{-(v/u)^2}$ with $u = \sqrt{2KT/m}$ where m is the mass of an atom, K is Boltzmann's constant, and T is the absolute temperature; the quantities G denote the coupling coefficients $G_i = \mu_i\varepsilon_i/\hbar$ and $G_3' = \mu_3\varepsilon_3'/\hbar$, while [22]

$$F^{(\pm)}(\mathbf{v}) = \frac{1}{\left\{\left(i\Delta_1^d + \Gamma_{10}\right)\left[i\left(\Delta_1^d + \Delta_2^d\right) + \Gamma_{20}\right] + |G_2|^2\right\}^2} \times \frac{1}{i\left(\Delta_1^d + \Delta_2^d \pm \Delta_3^d\right) + \Gamma_{30}}. \tag{3}$$

Here, $\Delta_i^d = \Delta_i + \mathbf{k}_i\cdot\mathbf{v}$ is the Doppler-shift frequency detuning and Γ_{n0} is the transverse relaxation rate between states $|n\rangle$ and $|0\rangle$. Let us consider the case in which beam 2 propagates along the direction of beam 3; then by setting $\mathbf{k}_1 = -k_1\hat{\mathbf{z}}$, $\mathbf{k}_2 = k_2\hat{\mathbf{z}}$, and $\mathbf{k}_3 = k_3\hat{\mathbf{z}}$, we have $\Delta_1^d = \Delta_1 - k_1 v$, $\Delta_2^d = \Delta_2 + k_1\zeta_2 v$, and $\Delta_3^d = \Delta_3 + k_1\zeta_3 v$, where $\zeta_2 = k_2/k_1$ and $\zeta_3 = k_3/k_1$ are the ratios between the magnitudes of the wave vectors.

Now, we investigate the AT splitting in the SWM spectrum in a Doppler-broadened system. We express $F^{(\pm)}$ in Eq. (3) as a function of v explicitly, then

$$F^{(\pm)}(v) = \frac{\eta^{(\pm)}}{[(v-\tilde{\Delta}_{10})(v-\tilde{\Delta}_{20}) - |\tilde{G}_2|^2]^2\left(v-\tilde{\Delta}_{30}^{(\pm)}\right)}. \tag{4}$$

Here, $\eta^{(\pm)} = 1/[k_1^3(1-\zeta_2)(1-\zeta_2\mp\zeta_3)]$, $\tilde{\Delta}_{10} = (\Delta_1 - i\Gamma_{10})/k_1$, $\tilde{\Delta}_{20} = (\Delta_1+\Delta_2-i\Gamma_{20})/[k_1(1-\zeta_2)]$, $\tilde{\Delta}_{30}^{(\pm)} = [\Delta_1+\Delta_2\pm(\Delta_3-i\Gamma_{30})]/[k_1(1-\zeta_2\mp\zeta_3)]$, and $|\tilde{G}_2|^2 = |G_2|^2/[k_1^2(1-\zeta_2)]$. By solving the pole structure in Eq. (4); namely,

$$(v-\tilde{\Delta}_{10})(v-\tilde{\Delta}_{20}) - |\tilde{G}_2|^2 = 0, \tag{5}$$

we obtain

$$F^{(\pm)}(v) = \frac{\eta^{(\pm)}}{(v-\tilde{v}_+)^2(v-\tilde{v}_-)^2\left(v-\tilde{v}_t^{(\pm)}\right)}. \tag{6}$$

Here,

$$\begin{aligned} \tilde{v}_\pm &= (1/2)(\tilde{\Delta}_{10}+\tilde{\Delta}_{20}) \pm (1/2)\sqrt{(\tilde{\Delta}_{10}-\tilde{\Delta}_{20})^2 + 4|\tilde{G}_2|^2}, \\ \tilde{v}_t^{(\pm)} &= \tilde{\Delta}_{30}^{(\pm)}. \end{aligned} \tag{7}$$

Therefore, we have

$$P^{(\pm)} \propto \int_{-\infty}^{\infty} dv \frac{e^{-(v/u)^2}}{(v-\tilde{v}_+)^2(v-\tilde{v}_-)^2\left(v-\tilde{v}_t^{(\pm)}\right)}, \tag{8}$$

and the SWM signal intensity is proportional to $|P^{(\pm)}|^2$.

We are interested in the SWM spectrum in a Doppler-broadened system. Let $v_\pm$ and $v_t^{(\pm)}$ be the real parts of $\tilde{v}_\pm$ and $\tilde{v}_t^{(\pm)}$, respectively, then the values of $v_\pm$ and $v_t^{(\pm)}$ vary as we scan the incident laser frequencies Δ_i, and the integral in Eq. (8) consists mainly of the contributions of atoms with velocities $v \simeq v_\pm$ and $v \simeq v_t^{(\pm)}$. For the case of $v_\pm, v_t^{(\pm)} \ll u$ then, due to the integration, the resonance of $P^{(\pm)}$ appears only when $v_+ = v_-$ or $v_\pm = v_t^{(\pm)}$. If we neglect the relaxation rates and define $\Delta_{10} = \Delta_1/k_1$, $\Delta_{20} = (\Delta_1+\Delta_2)/[k_1(1-\zeta_2)]$, and $\Delta_{30}^{(\pm)} = (\Delta_1+\Delta_2\pm\Delta_3)/[k_1(1-\zeta_2\mp\zeta_3)]$, then from $v_+ = v_-$ the resonant condition is

$$\Delta_{10} - \Delta_{20} = \pm\frac{2G_2}{k_1\sqrt{\zeta_2-1}}. \tag{9}$$

This equation is valid only when $\zeta_2 > 1$. On the other hand, for $v_\pm = v_t^{(\pm)}$ we have

$$\Delta_{30}^{(\pm)} = \frac{(\Delta_{10}+\Delta_{20}) \pm \sqrt{(\Delta_{10}-\Delta_{20})^2 + 4|\tilde{G}_2|^2}}{2}. \tag{10}$$

III. DRESSED-STATE MODEL

As is well known, in a homogeneously broadened system, AT splitting is observed when dressed states are resonantly excited by tuning the probe beam, thus the splitting equals the energy separation between the two dressed states. The situation is quite different in a Doppler-broadened system because, as we tune the laser frequencies, there will be atoms with specific velocities which can be in resonance with the dressed states through Doppler frequency shifting. One important consequence is that the AT spectrum depends strongly on the wave-number ratios, especially on the value of ζ_2. For example, according to Eq. (9), to observe the resonance originating from $v_+ = v_-$ it is required that $\zeta_2 > 1$. Also, in a cascaded four-level system, as Δ_3 is scanned the SWM spectrum exhibits a doublet structure only when $\zeta_2 < 1$ [22]. To reveal the underlying physics, in this section we shall employ the dressed-state model to study how the Doppler frequency shift affects the AT splitting. Through analyzing the frequency dependence of the resonant velocities, the features of the AT spectrum can be understood.

The strong coupling field from beam 2 which drives the transition between $|1\rangle$ and $|2\rangle$ creates dressed states. The corresponding Hamiltonian can be written as the matrix

$$H = \begin{pmatrix} \Delta_1^d & -G_2^* \\ -G_2 & \Delta_1^d + \Delta_2^d \end{pmatrix}, \tag{11}$$

from which the eigenenergies of the dressed states $|\pm\rangle$ are

$$\chi_\pm = \Delta_1^d + \tfrac{1}{2}\left[\Delta_2^d \pm \sqrt{\left(\Delta_2^d\right)^2 + 4|G_2|^2}\right]. \tag{12}$$

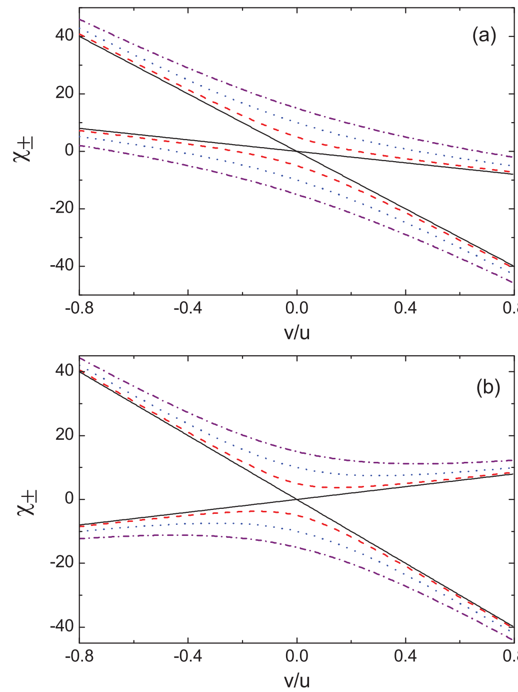

FIG. 2. (Color online) Dressed-state energy versus v with $\Delta_1/\Gamma_{30} = \Delta_2/\Gamma_{30} = 0$ and $\zeta_2 =$ (a) 0.8, (b) 1.2 for $G_2/\Gamma_{30} = 0$ (solid curve), 5 (dashed curve), 10 (dotted curve), and 15 (dash-dotted curve).

Due to the Doppler frequency shift, the energies of the dressed states depend on the atomic velocity. Specifically, we have

$$\chi_\pm(v) = -k_1 v\left(1-\frac{\zeta_2}{2}\right) + \left(\Delta_1 + \frac{\Delta_2}{2}\right) \pm \frac{1}{2}\sqrt{(\Delta_2 + k_1\zeta_2 v)^2 + 4|G_2|^2}. \tag{13}$$

Since the energy of the ground state $|0\rangle$ is 0, the resonance condition for the transition from the ground to the dressed state is $\chi_\pm(v) = 0$; thus, from Eq. (13), we obtain the resonant velocities

$$v_\pm = (1/2)(\Delta_{10}+\Delta_{20}) \pm (1/2)\sqrt{(\Delta_{10}-\Delta_{20})^2 + 4|\tilde{G}_2|^2}. \tag{14}$$

The above equation is exactly the same as Eq. (7) when relaxation rates are neglected. Physically, as a result of the Doppler frequency shift, atoms with velocities $v_\pm$ will be in resonance with the dressed states.

Let us first examine the velocity dependence of the dressed-state energy. Figure 2 presents the dressed-state energy $\chi_\pm$ versus v with $\Delta_1/\Gamma_{30} = \Delta_2/\Gamma_{30} = 0$, $G_2/\Gamma_{30} = 0$ (solid curve), 5 (dashed curve), 10 (dotted curve), and 15 (dash-dotted curve), while $\zeta_2 = 0.8$ for Fig. 2(a) and $\zeta_2 = 1.2$ for Fig. 2(b). In the absence of the coupling field (solid curves in Fig. 2), the dressed-state energies are just the frequency detunings for the one-photon and two-photon transitions; that is, $\Delta_1 - k_1 v$ and $\Delta_1 + \Delta_2 - k_1 v(1-\zeta_2)$, respectively. The two lines cross at $v = -\Delta_2/(k_1\zeta_2)$ and $\chi_+ = \chi_- = \Delta_1 + \Delta_2/\zeta_2$. On the other hand, the presence of the coupling field prevents level crossing, and the separation of the curves increases with the increase of G_2.

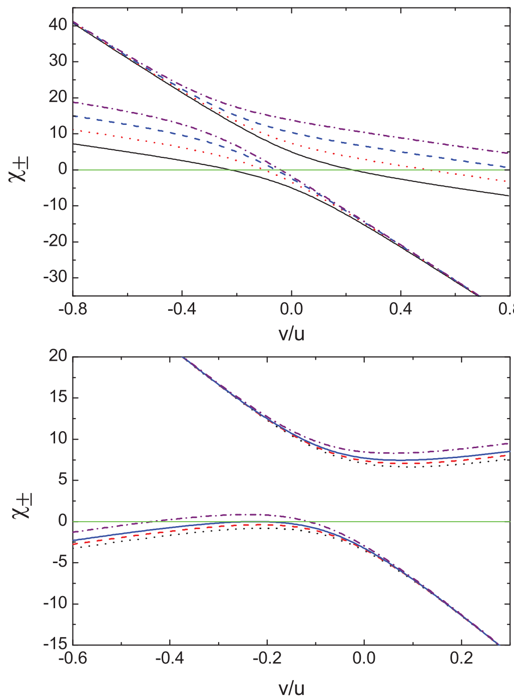

FIG. 3. (Color online) Dressed-state energy versus v with $\Delta_1/\Gamma_{30} = 0$, $G_2/\Gamma_{30} = 5$, and (a) $\zeta_2 = 0.8$, $\Delta_2/\Gamma_{30} = 0$ (solid curve), 4 (dotted curve), 8 (dashed curve), and 12 (dash-dotted curve); (b) $\zeta_2 = 1.2$, $\Delta_2/\Gamma_{30} = 3.5$ (dotted curve), 4 (dashed curve), 4.47 (solid curve), and 5.5 (dash-dotted curve). The resonant velocities $v_\pm$ correspond to the cross points of the curves $\chi_\pm(v)$ with a line $y = 0$ (thin solid line).

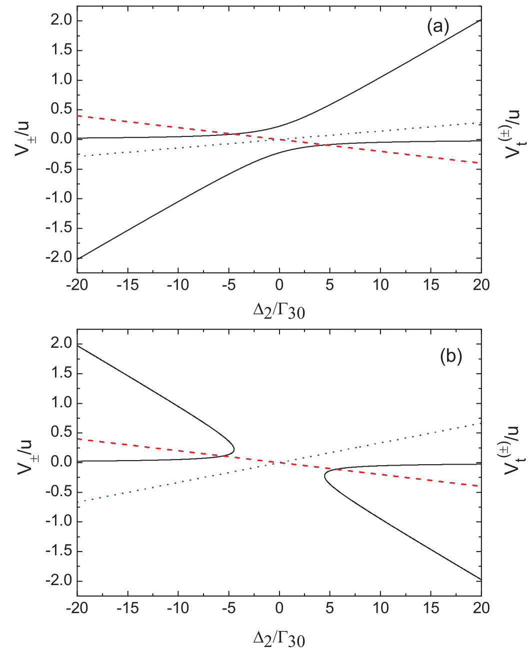

FIG. 4. (Color online) Resonant velocities $v_\pm$ (solid curve), $v_t^{(+)}$ (dashed curve), and $v_t^{(-)}$ (dotted curve) versus Δ_2 with $\Delta_1/\Gamma_{30} = \Delta_3/\Gamma_{30} = 0$, $G_2/\Gamma_{30} = 5$, and (a) $\zeta_2 = 0.8$, $\zeta_3 = 1.2$; (b) $\zeta_2 = 1.2$, $\zeta_3 = 0.8$.

We next study how the wave-number ratio ζ_2 affects $v_\pm$. Figure 3 presents $\chi_\pm$ versus v for $G_2/\Gamma_{30} = 5$, $\Delta_1/\Gamma_{30} = 0$, and [Fig. 3(a)] $\zeta_2 = 0.8$, $\Delta_2/\Gamma_{30} = 0$ (solid curve), 4 (dotted curve), 8 (dashed curve), and 12 (dash-dotted curve); [Fig. 3(b)] $\zeta_2 = 1.2$, $\Delta_2/\Gamma_{30} = 3.5$ (dotted curve), 4 (dashed curve), 4.47 (solid curve), and 5.5 (dash-dotted curve). The resonant velocities $v_\pm$ correspond to the crossing points of the curves $\chi_\pm(v)$ with a line $y = 0$ (thin solid line). When $\zeta_2 < 1$ [Fig. 3(a)], the velocities v_+ and v_- originate from the resonances of the dressed states $|+\rangle$ and $|-\rangle$, respectively. By contrast, we have both v_+ and v_- from the same dressed states when $\zeta_2 > 1$ [Fig. 3(b)]. This is because, in the absence of the coupling field, the dressed-state energy corresponding to the frequency detuning of the two-photon transition [i.e., $\Delta_1 + \Delta_2 - k_1 v(1 - \zeta_2)$] changes the sign of the slope as ζ_2 passes through 1 (see solid curves in Fig. 2). One important consequence is that the frequency-detuning dependence of the resonant velocities $v_\pm$ behaves in a completely different way for $\zeta_2 < 1$ and $\zeta_2 > 1$. Figure 4 presents the resonant velocities $v_\pm$ (solid curve), $v_t^{(+)}$ (dashed curve), and $v_t^{(-)}$ (dotted curve) as a function of the frequency detuning Δ_2/Γ_{30} when $G_2/\Gamma_{30} = 5$, $\Delta_1/\Gamma_{30} = \Delta_3/\Gamma_{30} = 0$, and [Fig. 4(a)] $\zeta_2 = 0.8$, $\zeta_3 = 1.2$; [Fig. 4(b)] $\zeta_2 = 1.2$, $\zeta_3 = 0.8$. As shown in Fig. 4(a), $v_\pm$ exists for any value of Δ_2 when $\zeta_2 < 1$. The solid curves of v_+ and v_- correspond to the velocities of atoms which are in resonance with the dressed states $|+\rangle$ and $|-\rangle$, respectively. On the other hand, there is a gap within which no $v_\pm$ exists when $\zeta_2 > 1$ [see Fig. 4(b)]. Specifically, from Eq. (14) there is no solution for $v_\pm$ in the regime $|\Delta_{10} - \Delta_{20}| < 2|G_2|/k_1\sqrt{\zeta_2 - 1}$. For example, the gap in Fig. 4(b) is $|\Delta_2| < 2\sqrt{\zeta_2 - 1}|G_2|$, or between -4.47 and 4.47. Moreover, the $v_\pm$ in the left regime originate from the resonance of the dressed state $|+\rangle$, while in the right regime they originate from $|-\rangle$.

Based on the dressed-state model, the main features of AT splitting in the SWM spectrum can be explained. Let us consider a cascade four-level system. As mentioned in Ref. [22], in the case of $\Delta_1 = \Delta_2 = 0$, the SWM spectrum exhibits a doublet structure as Δ_3 is scanned when $\zeta_2 < 1$; however, there is only a single peak in the SWM spectrum when $\zeta_2 > 1$ (see Fig. 2 in Ref. [22]). This can be explained by using the frequency-detuning dependence of the resonant velocities $v_\pm$. As shown in Fig. 4(a), $v_\pm$ exists for any value of Δ_2 when $\zeta_2 < 1$. These atoms can be resonant with the state $|3\rangle$ as Δ_3 is scanned, leading to a doublet structure in the spectrum. On the other hand, since there is no solution for $v_\pm$ in the gap $|\Delta_2| < 2\sqrt{\zeta_2 - 1}|G_2|$ when $\zeta_2 > 1$, there is only a single peak. Now, let us consider the spectrum when Δ_2 is scanned. As shown in Fig. 4, in this case both $v_\pm$ and $v_t^{(\pm)}$ vary. When $\zeta_2 < 1$ [Fig. 4(a)], the resonant condition $v_+ = v_-$ cannot be satisfied. In the cascade four-level system the resonance appears when the values of $v_\pm$ (solid curve) and $v_t^{(+)}$ (dashed curve) cross, leading to a doublet structure. On the other hand, since $v_\pm$ (solid curve) and $v_t^{(-)}$ (dotted curve) do not cross in a folded four-level system, there is only a single peak.

IV. MACROSCOPIC EFFECTS IN AT SPECTRUM

We have shown that, in a Doppler-broadened system, resonance appears when the condition $v_\pm = v_t^{(+)}$ ($v_\pm = v_t^{(-)}$) or $v_+ = v_-$ is satisfied. Previously, we studied the AT splitting corresponding to the resonant condition $v_\pm = v_t^{(+)}$ in a cascade four-level system [22], where atoms with velocity $v = v_\pm = v_t^{(+)}$ are doubly resonant with one of the dressed states and state $|3\rangle$. In this section, we shall study the AT splitting which originates from the condition $v_+ = v_-$ when $\zeta_2 > 1$. In contrast to the previous case, both v_+ and v_- correspond to the same dressed state when $\zeta_2 > 1$ [Fig. 3(b)], thus atoms with velocity $v = v_+ = v_-$ will only be resonant with one of the dressed states. Since there is a region for Δ_2 where atoms of two velocities v_+ and v_- are in resonance with the dressed state [see Fig. 4(b)] and so contribute to the SWM signal, a question arises concerning the nature of the resonant peaks corresponding to the condition $v_+ = v_-$. We shall show here that, in this situation, macroscopic effects play an important role in the formation of resonant peaks in the SWM spectrum.

Let us consider the case when Δ_2 is scanned and $\zeta_2 > 1$. If we set $\Delta_1 = \Delta_3 = 0$ then, according to Eq. (9), the resonant frequencies due to the condition $v_+ = v_-$ are $\Delta_2 = \pm 2\sqrt{\zeta_2 - 1}|G_2|$. In the following, we shall consider SWM in the cascade and folded four-level systems with parameters $\Gamma_{10}/\Gamma_{30} = \Gamma_{20}/\Gamma_{30} = 0.2$ and $\Gamma_{30}/(k_1 u) = 0.02$. Figure 5 presents the SWM intensity versus Δ_2 when $\zeta_2 = 1.2$, $\zeta_3 = 0.8$, $\Delta_1/\Gamma_{30} = \Delta_3/\Gamma_{30} = 0$, and $G_2/\Gamma_{30} = 0.1$ (solid curve), 5 (dashed curve), and 10 (dash-dotted curve) for [Fig. 5(a)] a cascade four-level system and [Fig. 5(b)] a folded four-level system. When the polarization interference is ignored, we obtain the dotted curve shown in Fig. 5(b) which is calculated from $\int_{-\infty}^{\infty} dv e^{-(v/u)^2} |\rho_{10}(r,v)|^2$ when $G_2/\Gamma_{30} = 5$. Let us consider first the case of a cascade four-level system. As shown

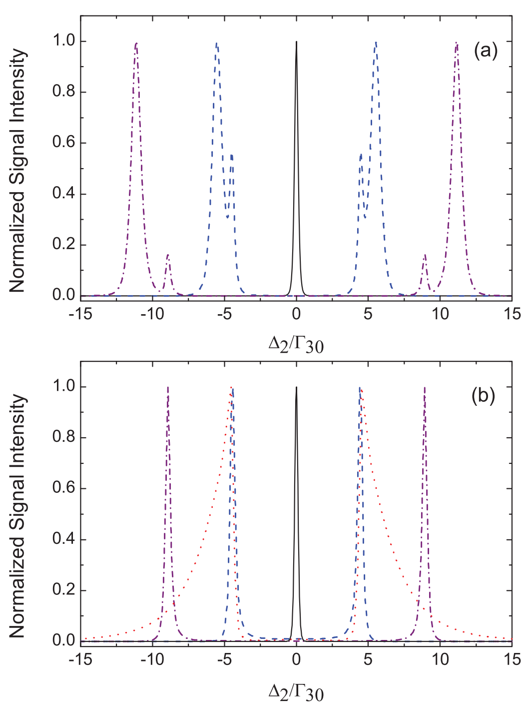

FIG. 5. (Color online) SWM intensity versus Δ_2 with $\Delta_1/\Gamma_{30} = \Delta_3/\Gamma_{30} = 0$, $\zeta_2 = 1.2$, $\zeta_3 = 0.8$, and $G_2/\Gamma_{30} = 0.1$ (solid curve), 5 (dashed curve), and 10 (dash-dotted curve) for (a) cascade and (b) folded four-level systems. The dotted curve in (b) is the spectrum when $G_2/\Gamma_{30} = 5$ and polarization interference is ignored.

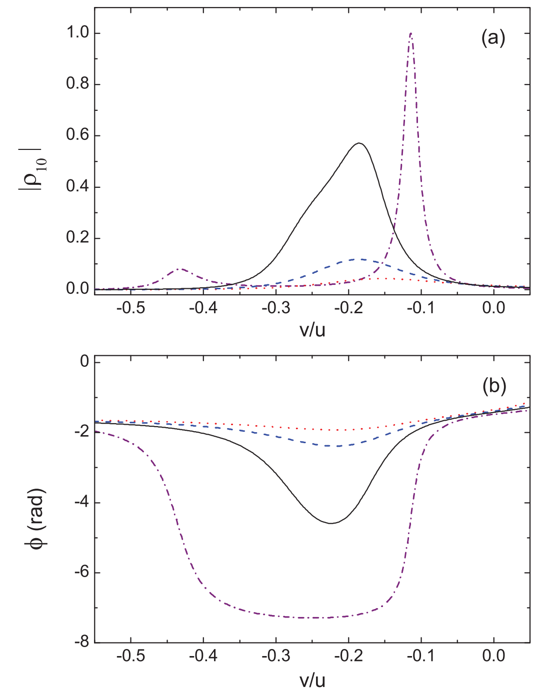

FIG. 6. (Color online) Velocity dependence of (a) $|\rho_{10}|$ and (b) ϕ, with $\Delta_1/\Gamma_{30} = \Delta_3/\Gamma_{30} = 0$, $\zeta_2 = 1.2$, $\zeta_3 = 0.8$, and $G_2/\Gamma_{30} = 5$, for $\Delta_2/\Gamma_{30} = 3.5$ (dotted curve), 4 (dashed curve), 4.47 (solid curve), and 5.5 (dash-dotted curve). In (a) the maximum of $|\rho_{10}|$ with $\Delta_2/\Gamma_{30} = 5.5$ is normalized to 1.

in Fig. 4(b), the values of $v_\pm$ (solid curve) and $v_t^{(+)}$ (dashed curve) cross; thus, the resonance derived from $v_+ = v_-$ is always accompanied by the resonance from $v_\pm = v_t^{(+)}$. In other words, two types of resonance appear simultaneously when $\zeta_2 > 1$ and the spectrum has four peaks [Fig. 5(a)]. We then consider the SWM spectrum in a folded four-level system. As shown in Fig. 4(b), $v_\pm$ (solid curve) and $v_t^{(-)}$ (dotted curve) do not cross, so the SWM spectrum exhibits a doublet structure [see Fig. 5(b)] due to the resonance originating from $v_+ = v_-$. The nature of this resonance can be clearly understood through a detailed analysis of the spectrum, since the resonance at $v_\pm = v_t^{(-)}$ is absent.

With the off-diagonal density matrix element expressed as $\rho_{10}(v) = |\rho_{10}|e^{i\phi}$, Fig. 6 presents [Fig 6(a)] $|\rho_{10}|$ and [Fig. 6(b)] ϕ versus v when $\zeta_2 = 1.2$, $\zeta_3 = 0.8$, $G_2/\Gamma_{30} = 5$, $\Delta_1/\Gamma_{30} = \Delta_3/\Gamma_{30} = 0$, and $\Delta_2/\Gamma_{30} = 3.5$ (dotted curve), 4 (dashed curve), 4.47 (solid curve), and 5.5 (dash-dotted curve). There exist three different regimes for the resonant velocities $v_\pm$ [Fig. 4(b)]. As shown in Fig. 3(b), there are no solutions for $v_\pm$ when $\Delta_2/\Gamma_{30} = 3.5$ (dotted curve) and 4 (dashed curve), thus all atoms are off resonance from the dressed states. The amplitude of the polarization increases as Δ_2 is tuned to the resonant frequency $2\sqrt{\zeta_2 - 1}|G_2|$. Since $\chi_-(v)$ is relatively flat near the point $v = v_+ = v_-$ [solid curve in Fig. 3(b)], atoms in a relatively wide region of velocity will contribute to the signal at resonance [solid curve in Fig. 6(a)]. On the other hand, atoms of velocities $v = v_+$ and $v = v_-$ will be in resonance with the dressed state when $\Delta_2/\Gamma_{30} = 5.5$ [dash-dotted curves in Fig. 3(b)]. As a result, there are two peaks in the velocity dependence of the polarization amplitude [dash-dotted curve in Fig. 6(a)]. These two peaks have narrower linewidths (i.e., a fewer number of atoms will contribute to SWM), causing a decrease in the signal. However, the change of the number of atoms involved cannot completely explain the sharp resonant peaks in the SWM spectrum. Specifically, if we neglect the polarization interference, then the SWM signal decreases slowly as Δ_2 is detuned from resonance [dotted curve in Fig. 5(b)]. Let us consider the velocity dependence of the phase of the polarization ϕ, as shown in Fig. 6(b). The phase ϕ varies sharply through about 2π near $v = v_-$ and $v = v_+$ (dash-dotted curve). As a result, destructive interference between the atom polarizations greatly suppresses the SWM. On the other hand, the interference is basically constructive near $v = v_+ = v_-$, leading to enhancement of the signal.

V. DISCUSSION AND CONCLUSION

In this paper, we have studied AT splitting in the SWM spectrum under the condition $v_+ = v_-$. This type of resonance is completely different in nature from the resonance originating from the condition $v_\pm = v_t^{(+)}$ studied in our previous paper [22]. Specifically, the condition $v_\pm = v_t^{(+)}$ corresponds to the case in which atoms with velocity $v = v_\pm = v_t^{(+)}$ have double resonance with one of the dressed states and with $|3\rangle$. By contrast, atoms with velocity $v = v_+ = v_-$ have only a single resonance with one of the dressed states. The appearance of this type of resonant peak in the SWM spectrum requires the involvement of macroscopic effects. As shown in Fig. 4(b), there are three different regimes as we scan Δ_2. At first, there is a gap within which no $v_\pm$ exists, so no atoms can be in resonance with the incident fields. Then, atoms with velocity $v = v_+ = v_-$ will be in resonance with one of the dressed states when Δ_2 is tuned to the frequency $\pm 2\sqrt{\zeta_2 - 1}|G_2|$, causing resonant enhancement of the SWM signal. Finally, there are two groups of atoms which are in resonance with the dressed states when Δ_2 is tuned to the regime with $v_+ \neq v_-$. However, besides the reduction in the number of the resonant atoms involved, destructive interference between the polarizations of atoms of different velocities greatly suppresses the SWM signal.

Based on the dressed-state model, we can explain the strong wave-number-ratio dependence of the AT splitting in the SWM spectrum. It is found that the characteristics of the resonant velocities $v_\pm$ depend strongly on the wave-number ratio ζ_2. As shown in Fig. 3, when $\zeta_2 < 1$, the velocities v_+ and v_- originate from the resonances of the dressed states $|+\rangle$ and $|-\rangle$, respectively. On the other hand, both v_+ and v_- originate from the same dressed state when $\zeta_2 > 1$. Correspondingly, $v_\pm$ exists for any value of Δ_2 when $\zeta_2 < 1$, while there is a gap within which no $v_\pm$ exists when $\zeta_2 > 1$ (Fig. 4). The underlying physics is that the two-photon detuning $\Delta_1^d + \Delta_2^d$ is independent of the velocity when $\zeta_2 = 1$, while its derivative [i.e, $d(\Delta_1^d + \Delta_2^d)/dv = -k_1(1 - \zeta_2)$] changes sign as ζ_2 passes through 1 (Fig. 2). Based on these features, the completely different SWM spectra for $\zeta_2 < 1$ and $\zeta_2 > 1$ presented in Ref. [22] and in this paper can be understood.

The condition $v_+ = v_-$ can also be employed to explain the EIT resonance in a Doppler-broadened three-level system.

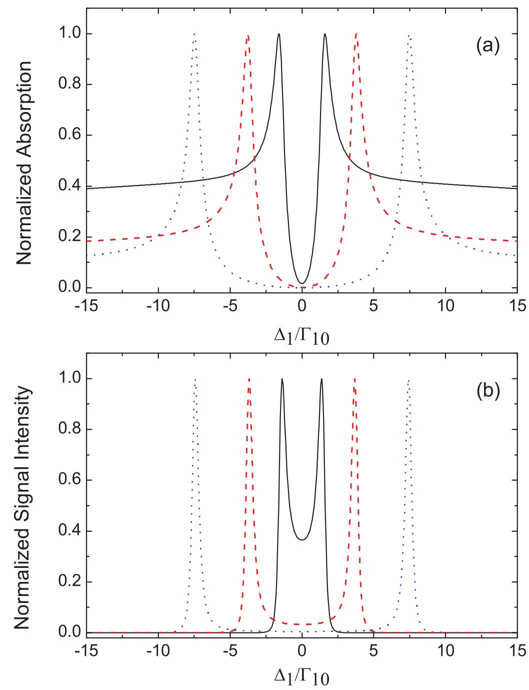

FIG. 7. (Color online) (a) Absorption of the EIT resonance and (b) SWM spectra against Δ_1 with $\Gamma_{10}/k_1u = 0.02$, $\Gamma_{20}/\Gamma_{10} = \Gamma_{30}/\Gamma_{10} = 0.2$, $\zeta_2 = 1.2$, $\zeta_3 = 0.8$, and $\Delta_2/\Gamma_{10} = \Delta_3/\Gamma_{10} = 0$, for $G_2/\Gamma_{10} = 2$ (solid curve), 5 (dashed curve), and 10 (dotted curve).

Let us consider a Doppler-broadened cascade $|0\rangle$-$|1\rangle$-$|2\rangle$ three-level system, where a strong coupling field couples the transition $|1\rangle$-$|2\rangle$, while a weak probe field is applied on the transition $|0\rangle$-$|1\rangle$. We are interested in the absorption of the probe beam in the presence of the coupling field. In a Doppler-broadened system the total susceptibility for the probe beam in the presence of a coupling field can be expressed as $\chi \propto i \int_{-\infty}^{\infty} d\mathbf{v} W(\mathbf{v}) F(\mathbf{v})$, where

$$F(\mathbf{v}) = \frac{i\left(\Delta_1^d + \Delta_2^d\right) + \Gamma_{20}}{\left(i\Delta_1^d + \Gamma_{10}\right)\left[i\left(\Delta_1^d + \Delta_2^d\right) + \Gamma_{20}\right] + |G_2|^2}, \quad (15)$$

and the absorption is proportional to the imaginary part of χ [30]. We consider the case of counterpropagating incident beams. Figure 7 presents [Fig. 7(a)] the absorption of the EIT resonance and [Fig. 7(b)] the SWM spectra versus Δ_1 in the folded four-level system, with the parameters $\Gamma_{10}/k_1u = 0.02$, $\Gamma_{20}/\Gamma_{10} = \Gamma_{30}/\Gamma_{10} = 0.2$, $\zeta_2 = 1.2$, $\zeta_3 = 0.8$, and $\Delta_2/\Gamma_{10} = \Delta_3/\Gamma_{10} = 0$, for $G_2/\Gamma_{10} = 2$ (solid curve), 5 (dashed curve), and 10 (dotted curve). Let us first consider the EIT resonance [Fig. 7(a)], where a transparency window appears in the absorption spectrum with a width that increases with the coupling-field intensity. On the other hand, there are two absorption peaks at the edges of the EIT window. If we compare Figs. 7(a) and 7(b), we find that the apparent coincidence of the peaks in the EIT resonance and SWM spectrum indicates that the absorption peaks in the former originates from the condition $v_+ = v_-$. A more detailed study of the effects of Doppler broadening on EIT is underway and will be presented later.

Finally, our dressed-state model can also be employed to study other types of high-resolution AT spectroscopy. For example, Ahmed and Lyyra [12] have analyzed the effects of Doppler broadening on AT splitting in the excitation spectra of a cascade $|0\rangle$-$|1\rangle$-$|2\rangle$ three-level system. They studied the fluorescence line shape from $|2\rangle$ and found that, in the case of counterpropagating incident beams, the Doppler-free AT doublet can be observed only when $\zeta_2 > 1$. According to our theory, this AT doublet originates from $v_+ = v_-$, and the resonant condition is given by Eq. (9). Some related experiments have been performed in sodium [13].

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