Vanishing and Revival of Resonance Raman Scattering

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The possibility to manipulate quantum coherence and interference, apart from its fundamental interest in quantum mechanics, is essential for controlling nonlinear optical processes such as high harmonic generation, multiphoton absorption, and stimulated Raman scattering. We show, analytically and numerically, how a nonlinear optical process via resonance Raman scattering (RRS) can be manipulated in a four-level double-Λ system by using pulsed laser fields. We find that two simultaneously excited RRS paths involved in the system can generate an ultimately destructive interference in the broad-bandwidth-limit regime. This, in turn, reduces the four-level system to an equivalent three-level system in a V configuration capable of naturally vanishing RRS effects. We further show that this counterintuitive phenomenon, i.e., the RRS vanishing, can be prevented by transferring a modulated phase of the laser pulse to the system at resonance frequencies. This work demonstrates a clear signature of both quantum destructive and constructive interference by actively controlling resonant multiphoton processes in multilevel quantum systems, and it therefore has potential applications in nonlinear optics, quantum control, and quantum information science.

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Since C. V. Raman first reported the Raman effect in 1928 [1], Raman spectroscopy has been widely used for characterizing the low-lying energy levels of atoms [2], molecules [3], and low-dimensional nanomaterials [4]. When the wavelength of the incident light falls within an absorption band of interest, the Raman scattering efficiency can be significantly enhanced via a resonant two-photon process. This leads to resonance Raman scattering (RRS), which is more selective than its non-resonance Raman counterpart [5–8]. A RRS process requires a three-level system in a Λ configuration with two low-lying energy levels of the ground electronic state, and an intermediate energy level in the excited electronic state. Under certain conditions, such a three-level Λ model can be selectively isolated from quantum systems with complex energy structures, e.g., by using narrow-bandwidth lasers, and has been used as a standard model for studying various types of nonlinear optical schemes, including coherent population trapping [9,10], electromagnetically induced transparency [11–15], and stimulated Raman adiabatic passage [16–20]. However, when a pulsed laser field is applied with a broad bandwidth over multiple off-resonant levels, more than one RRS path will be activated. Therefore the complexity will be dramatically increased owing to the nonlinear optical effect via quantum coherence and interference within and between paths, leading to many unexpected quantum interference phenomena. This, in turn, opens a new avenue for studying these nonlinear effects via multiple-optical-path quantum interference. Understanding how the nonlinear effect can be affected and, ultimately, manipulated by applied external fields remains a long-standing goal of both fundamental and practical significance in quantum science and technology [21–27].

Here, we theoretically examine a multiple-optical-path quantum interference in a typical four-level double-Λ (FLDL) system with two low-lying energy levels of the ground electronic state and two low-lying energy levels of the excited electronic state. When such a quantum system interacts with a transform-limited (TL) pulse with a sufficiently broad bandwidth, we show that two simultaneously excited RRS paths can destructively interfere with each other, leading to a natural vanishing of the RRS phenomenon. We further find that this RRS vanishing can be prevented by modulating the spectral phase of the laser pulse at resonance frequencies. These findings not only deepen our understanding of quantum interference but also demonstrate an active way of manipulating nonlinear
of an ultracold field $E$ interaction picture, with structure occurs, generating the hyperfine levels $2\mu$. For a narrow-bandwidth pulse $\mathcal{E}(t)$ can be given by [40,41]

$$
|\psi_{12m}(t)\rangle = \frac{\theta_{2n}(t)^{2} + \theta_{1m}(t)^{2}\cos[\theta_{12m}(t)]}{\theta_{12m}(t)^{2}}|1\rangle + \theta_{12m}(t)^{2}|2\rangle + \theta_{12m}(t)^{2}\cos[\theta_{12m}(t) - 1]|2\rangle + \frac{i\theta_{12m}(t)\sin[\theta(t)]}{\theta_{12m}(t)}|m\rangle,
$$

where the complex field areas $\theta_{1m}(t) = \mu_{1m} \int_{0}^{t} dt^{'\prime} \mathcal{E}(t^{'\prime})e^{-i\omega_{nm}t^{'\prime}}$, $\theta_{2m}(t) = \mu_{2m} \int_{0}^{t} dt^{'\prime} \mathcal{E}(t^{'\prime})e^{-i\omega_{nm}t^{'\prime}}$, and $\theta_{1m2}(t) = \sqrt[3]{(\theta_{1m}(t)^{2} + \theta_{2m}(t)^{2})}$, with $\omega_{nm} = (E_{m} - E_{n})$ and $\omega_{2m} = (E_{m} - E_{2})$. For a broad-bandwidth pulse with $\Delta\omega > \delta_{1}$, state $|2\rangle$ is accessible via RRS. For a narrow-bandwidth pulse with $\Delta\omega \ll \delta_{1}$, however, the three-level system reduces to a two-level system with $|\psi_{11m}(t)\rangle = \cos[\theta(t)]|1\rangle + i\sin[\theta(t)]|m\rangle$, i.e., $\theta(t) = \theta_{1m}(t)$ without the RRS to $|2\rangle$.

To show the dependence of the RRS on $\Delta\omega$, we consider $\mathcal{E}(t) = \text{Re}[(1/2\pi) \int_{0}^{t} A(\omega)e^{-i\delta(t)\omega\omega}d\omega]$, with spectral amplitude $A(\omega) = (A_{0}/\mu_{1m})\exp\left[-(\omega - \omega_{nm})^{2}/2(\Delta\omega)^{2}\right]$ to excite a three-level system of $|1\rangle$, $|2\rangle$, and $|3\rangle$. For this choice, the pulse area at $t_{f}$, i.e., $\theta_{1m}(t_{f}) \propto A(\omega_{1m})$, is independent of $\Delta\omega$ for a given $A_{0}$ at $\omega_{0} = \omega_{1m}$. As an example, we demonstrate such excitations in a three-level system of $|1\rangle$, $|2\rangle$, and $|3\rangle$, by excluding state $|4\rangle$, and numerically solve the corresponding equation (1) to calculate the wave function $|\psi_{123}(t)\rangle$. Figures 2(a) and 2(b) show a comparison of the numerically (exactly) calculated populations $P_{n}(t_{f}) = \langle n|\psi_{123}(t_{f})\rangle^{2}$ with the analytically derived populations $p_{n}^{0}(t_{f}) = \langle n|\psi_{123}^{(0)}(t_{f})\rangle^{2}$ ($n = 2, 3$) versus $\Delta\omega$. We choose $A_{0} = \pi/4$ and keep $\phi(\omega) = 0$, which, using Eq. (2), fix $\theta_{1m}(t_{f}) = \pi/4$ and lead to an equal population distribution between $|1\rangle$ and $|3\rangle$ in a narrow-bandwidth regime of $\Delta\omega \ll \delta_{1}$; see Figs. 2(a) and 2(b). The quantum state transfer (QST) to $|2\rangle$ appears in the three-level numerical (3LN) simulations as the bandwidth increases and asymptotically approaches a constant in the broad-bandwidth-limit regime, in good agreement with the three-level analytical (3LA) solutions.
We now focus on the FLDL system by considering the two closely spaced states $|3\rangle$ and $|4\rangle$ connected to $|1\rangle$ and $|2\rangle$. Figure 3(a) shows the dependence of $P_2(t_f) = |\langle 2 | \psi(t_f) \rangle|^2$ on $\Delta \omega$, for which the wave function $|\psi(t)\rangle$ is obtained by solving Eq. (1) with the FLDL model while using the same pulse as that in Fig. 2.

FIG. 2. The presence of resonance Raman scattering in a three-level $\Lambda$ system. (a),(b) Final populations $P_2(t_f)$ and $P_3(t_f)$ versus $\Delta \omega$ based on the three-level numerical (3LN) solutions at the resonant condition $\Delta = 0$, which are compared with the three-level analytical (3LA) solutions. (c),(d) The dependence of $P_2(t_f)$ and $P_3(t_f)$ on both $\Delta \omega$ and $\Delta$. The three-level $\Lambda$ system used in simulations is shown in (a).

Surprisingly, QST to $|2\rangle$ reaches a maximal value of 0.15% only around $\Delta \omega = 1.0 \delta_1$ and then decreases to a value of $< 10^{-4}$ in the broad-bandwidth-limit regime. Figure 3(b) shows the dependence of $P_2(t_f)$ on both $\Delta \omega$ and $\Delta$. The transition probability to $|2\rangle$ still remains extremely small, $< 10^{-4}$, even for a larger detuning. This implies that the RRS to $|2\rangle$ is significantly suppressed in the FLDL system.

We first analyze the pulse area theorem with Eq. (2) to qualitatively understand the underlying mechanism in Fig. 3. Since the energy splitting between $|3\rangle$ and $|4\rangle$ is extremely small, the transitions of $\Delta \omega$ and $\omega$ can be ignored in the limit regime of $\Delta \omega \gg \delta_1$, i.e., $A(\omega_{13}) \approx A(\omega_{14})$ and $A(\omega_{23}) \approx A(\omega_{24})$. This implies that the values of $\theta_{1m}(t_f)$ and $\theta_{2m}(t_f)$ are determined by the values of $\mu_{13}$, $\mu_{23}$, $\mu_{14}$, and $\mu_{24}$. According to Refs. [42,43], there is a geometrical structure of $\mu_{13} = -\sqrt{1/3} \mu_{14}$, $\mu_{23} = \mu_{14}$, and $\mu_{24} = \sqrt{1/3} \mu_{14}$, leading to the relation $\theta_{13}(t_f) \theta_{23}(t_f) \approx -\theta_{14}(t_f) \theta_{24}(t_f)$. When the complex pulse areas further satisfy the condition $|\theta_{13}(t_f)| \approx |\theta_{14}(t_f)|$, the two simultaneously excited RRS processes cancel each other out by using the TL pulse.

To further gain insights into the RRS vanishing in Fig. 3, we derive a pulse area theorem for the FLDL system driven by a pulsed field in the broad-bandwidth-limit regime (see the details in the Supplemental Material [44]). The time-dependent wave function of the system reads

$$|\psi^{(1)}(t)\rangle = \cos[\theta(t)]|1\rangle + \frac{i \theta_{13}(t)}{2 \theta(t)} \sin[\theta(t)]|3\rangle + \frac{i \sqrt{3} \theta_{13}(t)}{2 \theta(t)} \sin[\theta(t)]|4\rangle,$$

with $\theta_1(t) = 2 \theta_{13}(t)$ and $\theta(t) = |\theta_{13}(t)|$. The FLDL system is reduced into a three-level system in a $V$ configuration without QST to $|2\rangle$ at any time $t$, corresponding to the counterintuitive phenomenon of the RRS vanishing. Note that the derivation of Eq. (3) does not require the strict condition $|\theta_{13}(t_f)| \approx |\theta_{14}(t_f)|$ for generating the RRS vanishing. The analytical solution in Eq. (3) is valid only in the broad-bandwidth-limit regime. In the narrow-bandwidth regime, however, the two RRS paths will be naturally closed, corresponding to a three-level $V$ system, which has an analytical solution [40]

$$|\psi^{(1)}_{134}(t)\rangle = \cos[\theta(t)]|1\rangle + \frac{i \theta_{13}(t)}{\theta(t)} \sin[\theta(t)]|3\rangle + \frac{i \theta_{14}(t)}{\theta(t)} \sin[\theta(t)]|4\rangle$$

with $\theta(t) = \sqrt{|\theta_{13}(t)|^2 + |\theta_{14}(t)|^2}$. Clearly, the two three-level solutions by Eqs. (3) and (4) contain different physical meanings, but it is interesting that Eq. (4) is equivalent to
apply a Gaussian phase function pulse. In order to reduce the computational cost, we instead use a Gaussian phase modulation with a function \( \phi(\omega) = a \exp \left[-(\omega - \omega_c)^2/2b^2\right] \) centered at \( \omega_c = \omega_{13} \) with a width of \( b \) in the broad-bandwidth-limit regime, and of a Gaussian spectral amplitude \( A(\omega) \) (blue).

The RRS enhancements in Fig. 5(b) exhibit another maximum of \( P_2(t_f) = 0.4864 \) at \( a = \pi \) for \( b = 0.3\delta_2 \), indicating that the RRS can be manipulated by modulating the spectral phase at resonance frequencies. This enhancement approach differs from Refs. [43,47] by modulating the nonresonant spectral components of the pulses.

The RRS enhancements in Fig. 5(b) exhibit another maximum of \( P_2(t_f) = 0.4855 \) at \( a = 3.1\pi \) with a small shift from \( 3\pi \). All maximums become slightly higher than 0.4752 by directly inverting the sign of \( \mu_{13} \) and do not decrease to the minimum exactly at \( 2\pi \) and \( 4\pi \). These differences imply that the Gaussian phase with a width also modulates the spectral components around \( \omega_{13} \), leading to the interference of RRS with the detuned RRS. As a result, the RRS manipulation via this Gaussian phase modulation depends on the actual width of the phase function. A broad width of phase function with \( b = 0.5\delta_2 \) increases the enhancement in Fig. 5(b), indicating that the detuned-RRS paths play roles and constructively interfere with the RRS paths.

In summary, we theoretically examined a nonlinear optical effect via multiple-optical-path quantum interference in a FLDL \(^{87}\)Rb at ultracold temperatures. We found that a robust phenomenon of the RRS vanishing can be generated by using a TL pulse in the broad-bandwidth-limit regime.
regime. By transferring a modulated spectral phase of the laser pulse onto the system, we demonstrated that this counterintuitive phenomenon of the RRS vanishing could be prevented, leading to the RRS revival. This work provides a clear signature of both destructive and constructive interference toward ultimately manipulating resonant multiphoton optical processes in ultracold $^{87}$Rb atoms.

Ultracold $^{87}$Rb was the first and is the most popular atom for making Bose-Einstein condensates. This atom with excellent sensitivity is also studied for atomic clocks [48], quantum Raman memory [49], quantum sensors [50], quantum gate [51], and atom interferometers [52]. Our results contribute a new physical phenomenon to $^{87}$Rb and stimulated Raman scattering for exploiting nonlinear optical effects. The RRS vanishing within the present model requires the complex field areas to satisfy the condition

$$\theta_{13}(t_f)\theta_{23}^*(t_f) \approx -\theta_{14}(t_f)\theta_{24}^*(t_f)$$

in the broad-bandwidth-limit regime. According to Ref. [42], this condition is generally applicable to $^{87}$Rb (as well as to other alkali metal atoms) initially in a pure $m_F$ magnetic sublevel. It implies that the RRS vanishing also occurs for the system initially in a hyperfine level $F$ (in the absence of optical pumping). For a quantum system without the dipole relation as $^{87}$Rb atom, an optimized spectral phase could be designed to either suppress or enhance the RRS processes [53–56]. Since atomic and optical physics can also be demonstrated in artificial quantum systems such as superconducting circuits [57,58], and charged nitrogen or silicon vacancy center in diamond [59–61], we expect that this RRS manipulation can be applied to solid-state systems with potential applications to quantum information and quantum computing science [62–66].

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