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Quantum interference between Raman scattering and single-photon absorption*

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A new kind of quantum interference between Raman scattering and single-photon absorption is predicted theoretically, which gives an expanded view of quantum interference. Its potential application is also proposed.

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PACC: 4225, 0365, 0765, 7830

1. Introduction

Originally quantum interference was discovered as one of the critical features of photon, which testifies to the wave property of photon. The concept of interference was first clearly introduced in Young's double-slit experiment: a single light resource was split into two beams, when these two beams passed through two different paths and arrived at the same place, the light and shade stripes appeared on the screen. After that, the electron interference was realized by Davisson and Germer[1] according to the de Broglie hypothesis;[2] and recently the atom interference was identified[3] after Bose–Einstein condensation of atoms had been discovered.[4−6] Theoretically, any particle, no matter whether it is single or complex (such as Cooper pair[7] and photon pair[8]), has quantum interference, as long as its coherence length is long enough.

The quantum interference between single-photon absorption and multi-photon absorption in some certain systems was also proposed,[9−14] of which an example was the control of optical absorption of a gas by interference between one- and three-photon absorption pathways, generated by a fundamental optical pulse and a derived third harmonic optical pulse exciting the initial states to the final states, both of which were the same for the two pathways observed.[9] This new kind of two-path quantum interference effect is quite different from the interferences of particles, and it has many potential applications such as controlling the semiconductor exciton population[10] and producing asymmetric ionization of carriers in quantum well.[11]

In the present paper we propose a new quantum interference between two different processes: Raman scattering and single-photon absorption. It is known that whether it is one-and-two-photon interference[11] or one-and-three-photon interference,[13,15] they are both the interferences between two different transition paths in the same optical process, i.e. absorption. The interference between single-photon absorption and Raman scattering that we analyse below happens between two totally different processes: absorption and scattering, indicating a new kind of quantum interference. Apart from other applications, since this interference is related to the two very important optical processes, it may also have some potential applications in the analysis of the spectrum. In the structure analysis of materials, especially that of molecular materials, both single-photon absorption and Raman scattering are used as very important tools. However, sometimes in infrared and Raman spectra of noncentrosymmetrical materials, if two peaks are very close, it is quite impossible to indicate whether they are actually the same. In the following we show that the quantum interference between single-photon absorption and Raman scattering provides some new information, which may be used for solving this problem.

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2. Theoretical analysis

For simplicity, we consider here only two discrete states in a noncentrosymmetrical material shown in Fig.1, where $|i\rangle$, $|f\rangle$ and $|r\rangle$ represent the initial, the finial and the virtual states of a system, respectively. Three laser beams with frequencies ω_1, ω_2 and ω_3 focus on the material, and for simplicity again, the laser field is considered to interact with the energy level resonantly with $\Delta = \omega_1 = \omega_3 - \omega_2$, where Δ is the difference in energy between the initial and the finial states. The light loop is shown in Fig.2. At first, the two lasers with frequencies ω_2 and ω_3 are converged by a beam splitter, then the mix beam is split into two beams: one directly focuses on the sample and the other enters a differential frequency generator (DFG), and thereafter a beam with frequency ω_1 is produced, which also irradiates on the sample after a filter and a phase delay. Apparently, when the three lasers with the frequencies of ω_2, ω_2 and ω_3 interact with the system, two transition paths are available: Raman scattering and single-photon absorption (also shown in Fig.1).

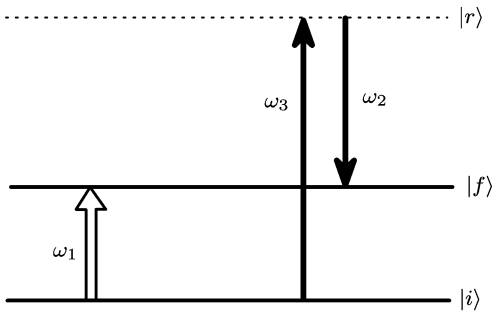


Fig.1. Raman Scattering and single-photon absorption in a two-level system.

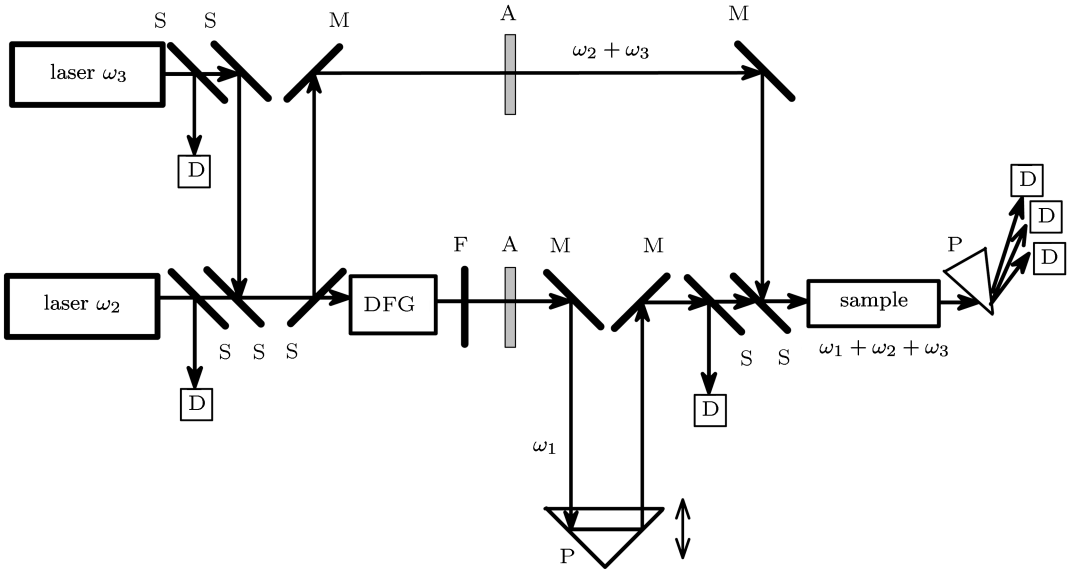


Fig.2. Quantum interference between Raman scattering and single-photon absorption, induced by two lasers and their difference frequency wave. DFG stands for the difference frequency generator, M denotes the reflecting mirror, S the beam splitter, D the detector, F the filter, A the attenuator, and P the prism.

As is known, the Hamiltonian of an electric dipole system, interacting with an external electromagnetic field, should be

$$H = H_0 - \boldsymbol{P} \cdot \boldsymbol{E},$$

where $\boldsymbol{P}$ is the polarization vector, H_0 the unperturbed Hamiltonian, the term $-\boldsymbol{P} \cdot \boldsymbol{E}$ is the perturbed contribution arising from the laser field. The vector $\boldsymbol{E}$ of a laser beam with a single harmonic component of frequency ω is considered as

$$\boldsymbol{E}(t) = \boldsymbol{E}_\omega \mathrm{e}^{-\mathrm{i}\,\omega t} + \boldsymbol{E}_\omega^* \mathrm{e}^{\mathrm{i}\,\omega t},$$

where the quantity marked with the asterisk is a complex conjugate.

Considering the single-photon absorption only, the system transits from $|i\rangle$ to $|f\rangle$ by absorbing a photon with frequency ω_1. The transition amplitude is

$$a_{\rm S}(t) = -\boldsymbol{P}_{fi}\cdot\boldsymbol{E}_{\omega_1}\frac{{\rm e}^{-{\rm i}(\hbar\omega_1-E_f+E_i)t/\hbar}}{\hbar\omega_1-E_f+E_i},$$

where transition dipole $\boldsymbol{P}_{fi}=\langle f|\boldsymbol{P}|i\rangle$, for simplicity we set

$$A=-\boldsymbol{P}_{fi}\cdot\boldsymbol{E}_{\omega_1},$$

and rewrite the transition amplitude of single-photon absorption as

$$a_{\rm S}(t) = A\frac{{\rm e}^{-{\rm i}(\hbar\omega_1-E_f+E_i)t/\hbar}}{\hbar\omega_1-E_f+E_i}.$$

The single-photon absorption rate from $|i\rangle$ to $|f\rangle$ is given by

$$\begin{aligned}T_{\rm S}(\omega_1) &= \frac{\rm d}{{\rm d}t}\left|a_{\rm S}(t)\right|^2_{t=0}\\ &= 2\pi\left|A\right|^2\delta(\hbar\omega_1-E_f+E_i).\end{aligned}$$

Raman scattering also happens in this system by absorbing a photon with ω_3 and emitting a photon with ω_2. However, since two lasers with ω_3 and ω_2 are involved at the same time, the Raman scattering here is different from the Raman scattering seen usually in which only the laser with ω_3 is involved. It is rather a "stimulated Raman scattering (SRS)" with a much bigger transition rate than usual. Its transition amplitude from the second-order perturbation calculation is given by

$$a_{\rm R}(t) = B\frac{{\rm e}^{-{\rm i}(\hbar\omega_3-\hbar\omega_2-E_f+E_i)t/\hbar}}{\hbar\omega_3-\hbar\omega_2-E_f+E_i},$$

where

$$\begin{aligned}B=\sum_r\Bigg[&\frac{(\boldsymbol{P}_{fr}\cdot\boldsymbol{E}^*_{\omega_2})(\boldsymbol{P}_{ri}\cdot\boldsymbol{E}_{\omega_3})}{E_r-E_i-\hbar\omega_3}\\ &+\frac{(\boldsymbol{P}_{ir}\cdot\boldsymbol{E}^*_{\omega_2})(\boldsymbol{P}_{rf}\cdot\boldsymbol{E}_{\omega_3})}{E_i-E_r+\hbar\omega_2}\Bigg],\end{aligned}$$

and the transition rate is

$$\begin{aligned}T_{\rm R}(\omega_2,\omega_3) &= \frac{\rm d}{{\rm d}t}\left|a_{\rm R}(t)\right|^2_{t=0}\\ &= 2\pi\left|B\right|^2\delta(\hbar\omega_3-\hbar\omega_2-E_f+E_i).\end{aligned}$$

When these two transition paths from the same initial and the same final states exist at the same time, it is impossible to determine whether the transition of the system from $|i\rangle$ to $|f\rangle$ is via single-photon absorption or Raman scattering, for there is a quantum interference between them. Therefore we cannot simply add the two transition rates together to obtain the total transition rate. First, we need the total transition amplitude, which is

$$a(t) = a_{\rm S}(t)+a_{\rm R}(t),$$

and then the total transition rate is

$$\begin{aligned}T &= \frac{\rm d}{{\rm d}t}\left[\left|a_{\rm S}\right|^2+\left|a_{\rm R}\right|^2+a^*_{\rm S}a_{\rm R}+a^*_{\rm R}a_{\rm S}\right]_{t=0}\\ &= 2\pi(|A|^2+|B|^2+(A^*B+AB^*))\\ &\quad\times\delta(\hbar\omega_1-E_f+E_i).\end{aligned}$$

Using $E_\omega=|E_\omega|{\rm e}^{\phi}$, we can obtain

$$\begin{aligned}T = 2\pi(&|A|^2+|B|^2\\ &+2|A||B|\cos(\phi_{\omega_1}+\phi_{\omega_2}-\phi_{\omega_3}))\\ &\times\delta(\hbar\omega_1-E_f+E_i),\end{aligned}$$

i.e.

$$T = T_{\rm S}+T_{\rm R}+2\sqrt{T_{\rm S}T_{\rm R}}\cos(\phi_{\omega_1}+\phi_{\omega_2}-\phi_{\omega_3}), \quad (1)$$

which explicitly shows how the interference contribution is related to the phases of the three optical beams. According to this result, if we select appropriate intensities of the incident beams with the single-photon absorption amplitude and the Raman scattering amplitude matching each other, and then, if we tune the phase term to $2n\pi$ (n is an integer), the total transition will reach the maximum, or if we tune the phase term to $2n\pi+\pi$, the total transition will become zero. Therefore, by tuning the phase term, we can obtain a complete destructive interference between Raman scattering and single-photon absorption, which makes neither Raman scattering nor single-photon absorption take place in the material.

The experimental setup shown in Fig.2 can be further simplified by using only two lasers with $\Delta=\omega=2\omega-\omega$ in some cases. The sketch map of the light loop is shown in Fig.3. At first the laser with frequency ω is split into two beams by a beam splitter, then, the frequency of one subsequent beam is doubled through a second harmonic generator (SHG), finally, after a filter and a phase delay, the beam of frequency 2ω is combined with the beam of frequency ω by another splitter. Based on the result of the general quantum interference that we have presented above, the interference term of this process is given by

$$T = T_{\rm S}+T_{\rm R}+2\sqrt{T_{\rm S}T_{\rm R}}\cos(2\phi_\omega-\phi_{2\omega}). \quad (2)$$

The simplified setup is similar to that in the experiments on interference between one-and-two-photon absorption pathways.[12] It may have some advantages over the setup shown in Fig.2 in some cases, but the second harmonic generation is difficult to achieve when ω is in the far infrared region, which restricts the application of the simplified setup.

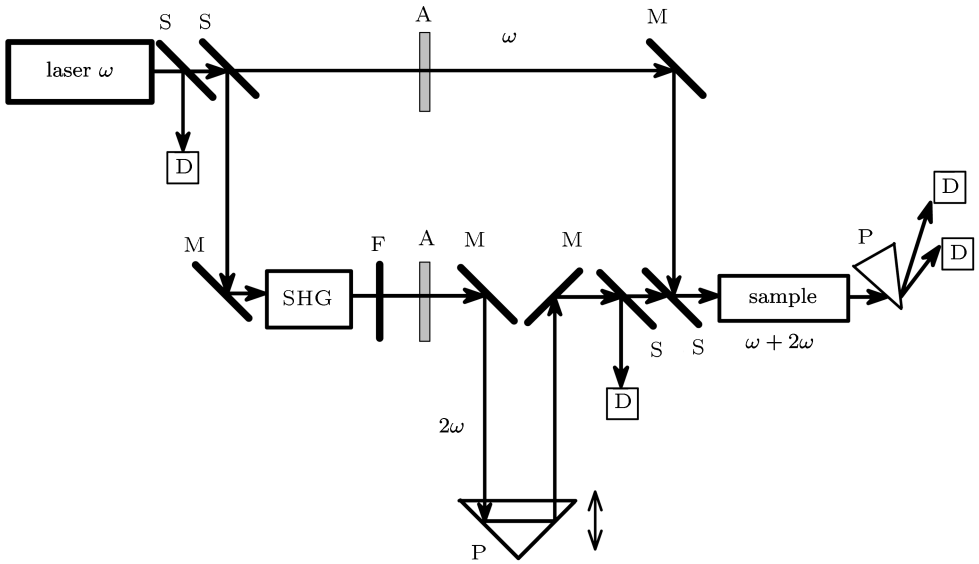


Fig.3. Quantum interference between Raman scattering and single-photon absorption, induced by a laser and its second harmonic wave.

3. Application

The quantum interference between single-photon absorption and stimulated Raman scattering has the same potential applications as the quantum interference between single-photon and multi-photons. Besides, this new type of quantum interference between optical transitions may also have the following advantages: (i) in most cases, stimulated Raman scattering is much more efficient than multi-photons absorption, while the efficiency is known to be crucial to all applications; (ii) in the far infrared region, especially the terahertz region, the harmonic generation of the laser beam is hard to achieve, and the quantum interference between single-photon absorption and multi-photons absorption cannot be realized by the technique available currently. If we utilize the quantum interference between single-photon absorption and stimulated Raman scattering, the laser ω_2 and the laser ω_3 could be in the visible and near infrared region where high-power tunable lasers are available, which makes the job a lot more easier.

For a material with a centrosymmetrical structure, some transitions display solely infrared activity while other transitions exhibit only Raman activity, and hence it produces a reciprocal Raman or infrared spectrum. For materials with a noncentrosymmetrical structure, some transitions may exhibit both infrared and Raman activities, which can be detected in both Raman and infrared spectra. Therefore, from the Raman and infrared spectra measured from a material, we can know whether it has centrosymmetry. However, when we consider the two transitions in a noncentrosymmetrical material (one is from infrared absorption and the other is from Raman scattering), which have the same frequencies, yet, it used to be very hard to indicate whether they were from the same initial to final states. Now that the theoretical analysis is presented in the present article, it is no problem to do so any more. Since the complete destructive interference between Raman scattering and single-photon absorption happens only when the two transitions take place from the same initial state to the same final state, therefore, the initial and final states of the two transitions should be the same if the complete destructive interference is observed; otherwise, they should be different. Obviously, understanding the transitions will provide an additional important clue to the analysis of material structures.

4. Summary

We have theoretically analyzed the interference between Raman scattering and single-photon absorption, induced by using phase-correlative laser beams with different frequencies. We have also proposed its possible applications, where more information about molecule structure and electronic state may be disclosed.

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