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Abstract

Femtosecond coherent anti-Stokes Raman scattering (CARS) spectra suffer from low spectral resolution because of the broadband laser spectrum. In this paper, we propose a feasible scheme to achieve a high-resolution two-pulse CARS spectrum by shaping both the pump and probe pulses using rectangular amplitude modulation. We show that a narrowband hole in the CARS spectrum can be created by the amplitude-shaped laser pulse, the position of which is correlated with the Raman resonant frequency of the molecule. Thus, by observing holes in the CARS spectrum, we are able to obtain a high-resolution CARS spectrum and the energy-level diagram of the molecule.

(Some figures may appear in colour only in the online journal)
we propose rectangular spectral amplitude modulation to improve the spectral resolution of two-pulse CARS. In the two-pulse CARS process [19, 20], the pump and probe pulses share the same laser field, as shown in figure 1(b). Compared with single-pulse or three-pulse CARS [13–18], two-pulse CARS uses simpler experimental apparatus and has wider practical applications. Our results show that a narrowband CARS uses simpler experimental apparatus and has wider spectral resolution of CARS [13–17], involving CARS gating, has been proved to be a tool to improve the spectral resolution of CARS [13–17], CARS spectrum narrowing and CARS background suppression. However, here we propose a form of spectral amplitude modulation with rectangular tailoring of both the pump and probe laser spectra to improve the spectral resolution of two-pulse CARS. This simple amplitude modulation is shown in figure 1(c), where δ and Δ represent the amplitude step position and amplitude modulation width, respectively. By this rectangular amplitude modulation, the frequency components in the rectangular window are cut off while those outside the window are preserved. In our simulation, the laser central frequencies of the pump, Stokes and probe pulses are set to be ωp = 13 500 cm^{-1}, ωs = 12 500 cm^{-1} and ωpr = ωp = 13 500 cm^{-1}, and their spectral bandwidths (full width at half maximum) are all 200 cm^{-1}. We first demonstrate control of the CARS spectrum in benzene with a Raman level of 992 cm^{-1}. Figure 2 presents.
Figure 2. CARS spectra in benzene induced by a shaped laser pulse with an amplitude step position $\delta = 13500 \text{ cm}^{-1}$ for amplitude modulation widths $\Delta = 20$ (black solid line), 60 (red dashed line) and 100 cm$^{-1}$ (blue dotted line).

Figure 3 shows CARS spectra in benzene induced by a shaped laser pulse with an amplitude step position $\delta = 13500 \text{ cm}^{-1}$ for amplitude modulation widths $\Delta = 20$ cm$^{-1}$ (black solid line), 13500 cm$^{-1}$ (red dashed line) and 13560 cm$^{-1}$ (blue dotted line). As can be seen, in each case the CARS spectrum is strongly modulated, and a hole in the CARS spectrum is created. With increasing amplitude modulation width $\Delta$, the CARS intensity decreases and the width of the hole increases. Thus, by applying a small amplitude modulation width $\Delta$, a narrowband hole in the CARS spectrum can be obtained. The width of the hole (full width at half depth) is only $\sim 24 \text{ cm}^{-1}$ for an amplitude modulation width $\Delta = 20 \text{ cm}^{-1}$ (see the black solid line in figure 2).

Figure 3. The CARS spectra in benzene induced by a shaped laser pulse with an amplitude modulation width $\Delta = 20 \text{ cm}^{-1}$ for amplitude step positions $\delta = 13440$ (black solid line), 13500 (red dashed line) and 13560 cm$^{-1}$ (blue dotted line).

Figure 4. CARS spectrum in pyridine induced by an unshaped laser pulse (black dashed line) and a shaped laser pulse with an amplitude modulation width $\Delta = 20 \text{ cm}^{-1}$ and an amplitude step position $\delta = 13500 \text{ cm}^{-1}$ (red solid line). Only a single broadband CARS signal is observed for the unshaped laser pulse, while two distinct holes in the CARS spectrum are observed at 14488 and 14528 cm$^{-1}$ for the amplitude-shaped laser pulse. Based on the relation $\omega_{\text{as-hole}} = \delta + \Omega_R$, it is easy to verify that the two holes are related to the two Raman levels of the pyridine. Therefore, by observing the two holes in the CARS spectrum, a high-resolution CARS spectrum can be obtained—this result is similar to that obtained by spectral notch shaping in single-pulse CARS [18], whereas it is different from that obtained by $\pi$ phase modulation in three-pulse CARS [13], where the CARS spectrum is greatly narrowed.

The high spectral resolution of the Raman levels can also be obtained by shifting the rectangular amplitude step position.
and measuring the CARS intensity at a given frequency. Figure 5 presents the CARS intensity at a frequency $\omega_{as} = 14500 \text{ cm}^{-1}$ in benzene (a) and pyridine (b) as a function of the amplitude step position $\delta$ for an amplitude modulation width $\Delta = 20 \text{ cm}^{-1}$. As can be seen, one hole at an amplitude step position $\delta = 13508 \text{ cm}^{-1}$ is observed in benzene, while two distinct holes at amplitude step positions $\delta = 13512$ and $13472 \text{ cm}^{-1}$ are observed in pyridine. By the relation $\omega_{as}$-hole $= \delta + \Omega_R$, the Raman resonant frequencies can be calculated as $\Omega_R = 992 \text{ cm}^{-1}$ in benzene and $\Omega_R = 988$ and $1028 \text{ cm}^{-1}$ in pyridine—in agreement with the known Raman resonant frequencies of benzene and pyridine mentioned above.

In conclusion, we have theoretically demonstrated that two-pulse CARS spectra can be modulated by shaping both the pump and probe pulses using rectangular amplitude modulation. It was shown that amplitude-shaped laser pulses can induce a narrowband hole in the CARS spectrum, the position of which is correlated with the Raman resonant frequency of the molecule. It was also shown that a high-resolution CARS spectrum and an energy-level diagram of the molecule can be obtained by observing the holes in the CARS spectrum. We believe that these theoretical results appear promising for applications in microscopic investigations into complex molecular structures and various related fields.

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References