

Spectral-focusing coherent anti-Stokes Raman scattering driven by bright correlated twin beams

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An understanding of the interaction between bright correlated twin beams and matter is important for expanding the applications of optical quantum technology. In particular, it is worthwhile to clarify the advantages of twin-beam correlation in spectroscopic applications, since twin-beam sources that use parametric down-conversion have a wide range of wavelength tunability. Here, we theoretically and experimentally demonstrate that the spectral-focusing technique used in coherent anti-Stokes Raman scattering (CARS) can be implemented by twin beams without the use of ultrashort-pulse lasers. In typical spectral focusing using ultrashort pulses, spectral resolution is improved by narrowing the instantaneous frequency width due to frequency chirping. Similarly, we show theoretically that the formation of a chirp structure within the temporal correlation of twin beams produces an effect similar to typical spectral focusing. To experimentally demonstrate twin-beam spectral focusing, CARS spectroscopy of polyethylene is performed when the time delay between the chirped twin beams is set within the correlation width or it is set outside the correlation width. We observe that the spectral resolution is improved when the correlation is effective. By comparing with theoretical simulations, we confirm that the improvement is caused by frequency chirping.

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I. INTRODUCTION

Bright correlated twin beams generated by high-gain parametric down-conversion (PDC) have attracted attention for their ability to produce ultrafast temporal correlations [1–4]. These correlations are formed through a mechanism similar to the generation of an ultrashort optical pulse, which relies on phase synchronization over a broad bandwidth. Specifically, the counterpart of the classical electric field phase is the phase sum of the signal and idler modes, the synchronization of which creates ultrafast temporal correlation. This similarity suggests that unique techniques based on spectral-phase-controlled ultrashort pulses, such as spectral focusing [5], temporal focusing [6,7], and time stretching [8], used in optical spectroscopy

and imaging applications, could be realized using twin beams. Furthermore, the PDC process used to generate twin beams allows for a wide range of wavelength tuning [9]. Therefore, if the unique capabilities of ultrashort pulses in spectroscopic measurements can be achieved with twin-beam light sources, twin beams could become powerful optical probes that combine broad tunability with the advantages of ultrashort pulses.

An application that is likely to be highly compatible with twin-beam light sources is spectral-focusing nonlinear Raman-scattering spectroscopy [5]. To induce nonlinear Raman processes such as stimulated Raman scattering [10] and coherent anti-Stokes Raman scattering (CARS) [11], it is necessary to make the beat frequency of the two-color intense light beams resonantly match the Raman transition. Since a PDC light source in the high-gain regime can generate two-color intense beams with a variable frequency difference, CARS can be induced using only a PDC light source [12]. In the spectral-focusing technique that uses ultrashort pulses, the narrowing of the instantaneous frequency width induced by frequency chirping is harnessed to improve the spectral resolution [5]. The recent

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observation of frequency chirping within the ultranarrow temporal correlation of twin beams [13] suggests that the spectral-focusing technique is also feasible for twin beams.

In this work, CARS spectroscopy driven by bright twin beams is theoretically and experimentally investigated. By extending the theoretical model of four-wave mixing given in Ref. [13], we derive the model of CARS driven by twin beams. It is shown theoretically that the spectral-focusing technique can be realized by using twin beams with frequency chirping within the temporal correlation.

In the experiments, twin beams are generated by high-gain PDC with nanosecond-pulsed excitation. As a prelude to investigating the effect of twin-beam correlation, the correlation properties are evaluated by cross-correlation measurement with a nonresonant CARS (NCARS) process, confirming that the phase dispersion generates a chirp structure within the temporal correlation. To investigate the effect of twin-beam correlation on the CARS spectra, we compare two types of CARS spectroscopy of polyethylene, namely, CARS spectroscopy using the uncorrelated twin beams and CARS spectroscopy using the correlated twin beams. CARS spectroscopy using the uncorrelated twin beams is performed under conditions in which the correlation is effectively canceled by adjusting the delay between the twin beams outside the temporal-correlation width. The two large Raman spectral peaks for polyethylene, separated by 35 cm^{-1} , cannot be distinguished in the uncorrelated twin-beam CARS spectrum. In twin-beam correlation CARS spectroscopy, the spectra of the twin-beam enhanced CARS light are measured by varying the delay within the temporal correlation width of hundreds of femtoseconds. In this case, the two Raman spectral peaks for polyethylene can be distinguished by setting the appropriate delay. This result shows improved spectral resolution compared to CARS using uncorrelated twin beams. By performing numerical simulations with and without phase dispersion for twin beams, we confirm that this improvement in spectral resolution is caused by frequency chirping.

II. THEORETICAL STUDY OF COHERENT ANTI-STOKES RAMAN SCATTERING (CARS)

A. Procedure of CARS spectroscopy using twin beams

Before going into the details of the theoretical model, we briefly explain the procedure for CARS spectroscopy using twin beams. In Fig. 1, we show the experimental setup for twin-beam generation and CARS driven by twin beams. The intense signal and idler twin beams are produced by type-0 PDC process. After a variable time delay τ is added to the idler beam, the beams are focused and incident on the sample, and the generated CARS light is detected. In the following, we will derive the theoretical model for this spectroscopic method.

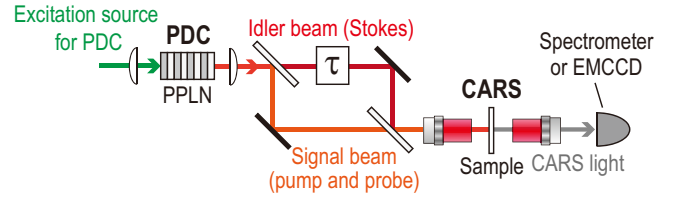


FIG. 1. The experimental setup for CARS spectroscopy using twin beams.

B. Derivation of CARS light spectra driven by twin beams

We introduce a theoretical model for CARS driven by twin beams to study the effect of twin-beam correlation by extending the theoretical model derived in Ref. [13]. The annihilation operator for twin beams generated by type-0 PDC can be expressed using the vacuum-field operators $\hat{a}_{\text{vac}}(\omega)$ and $\hat{a}_{\text{vac}}^\dagger(\omega)$ as $\hat{a}(\omega) = [\mu(\omega)\hat{a}_{\text{vac}}(\omega) + i\nu(\omega)\hat{a}_{\text{vac}}^\dagger(2\Omega - \omega)]$, where $\mu(\omega)$ and $\nu(\omega)$ are the parametric interaction propagation factors and 2Ω is the angular frequency of the monochromatic excitation source for PDC. $\mu(\omega) = \mu(2\Omega - \omega)$ and $\nu(\omega) = \nu(2\Omega - \omega)$ are satisfied for a type-0 process [14].

The field operator at frequency $\omega_C = \omega_{\text{pu}} - \omega_{\text{st}} + \omega_{\text{pr}}$ for CARS light can be expressed in the following form:

$$\begin{aligned} \hat{a}_{\text{CARS}}(\omega_C, \tau) &= \int_{\Omega}^{2\Omega} d\omega_{\text{pr}} \int_0^{\Omega} d\omega_{\text{st}} \int_{\Omega}^{2\Omega} d\omega_{\text{pu}} \\ &\times e^{-i\omega_{\text{st}}\tau} \chi^{(3)}(\omega_{\text{pu}} - \omega_{\text{st}}) \eta(\omega_{\text{pr}}) \eta^*(\omega_{\text{st}}) \eta(\omega_{\text{pu}}) \\ &\times \hat{a}(\omega_{\text{pr}}) \hat{a}^\dagger(\omega_{\text{st}}) \hat{a}(\omega_{\text{pu}}) \delta(\omega_{\text{pu}} - \omega_{\text{st}} + \omega_{\text{pr}} - \omega_C), \end{aligned} \quad (1)$$

where the subscripts pu, st, and pr represent the pump, Stokes, and probe fields, respectively, as shown in the energy diagram in Fig. 2. By focusing on each integration interval of Eq. (1), it can be seen that the signal beam plays the role of the pump and probe and the idler

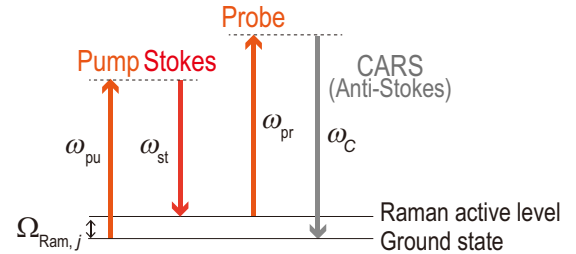


FIG. 2. The energy diagram for CARS. The CARS light (anti-Stokes light) is produced when the frequency difference between the pump light and the Stokes light is equal to the molecular resonance frequency ($\Omega_{\text{Ram},j}$).

beam plays the role of the Stokes beam. The exponential factor ($e^{-i\omega_{st}\tau}$) is the frequency-dependent phase factor added by the delay of the idler beam. $\eta(\omega)$ represents the accumulated phase induced by the optical elements and spatial diffraction [2]. $\chi^{(3)}(\omega)$ is expressed as the sum of the nonlinear resonant and NCARS susceptibilities,

namely, $\chi^{(3)}(\omega) = \sum_j \chi_{R,j}^{(3)}(\omega) + \chi_{NR}^{(3)}$, where $\chi_{R,j}^{(3)}(\omega) \propto 1/[(\Omega_{Ram,j} - \omega) - i\Gamma_{Ram,j}]$ is the complex Lorentz function with Raman resonance frequency $\Omega_{Ram,j}$ and line width $\Gamma_{Ram,j}$, and $\chi_{NR}^{(3)}$ is set to a constant value. The CARS spectrum can be written as

$$\begin{aligned}
 S(\omega_C, \tau) &= \langle 0 | \hat{a}_{CARS}^\dagger(\omega_C, \tau) \hat{a}_{CARS}(\omega_C, \tau) | 0 \rangle \\
 &= \int_{\Omega}^{2\Omega} d\omega'_{pu} \int_0^{\Omega} d\omega'_{st} \int_{\Omega}^{2\Omega} d\omega'_{pr} \int_{\Omega}^{2\Omega} d\omega_{pr} \int_0^{\Omega} d\omega_{st} \int_{\Omega}^{2\Omega} d\omega_{pu} \\
 &\quad \times e^{i(\omega'_{st} - \omega_{st})\tau} \chi^{(3)*}(\omega'_{pu} - \omega'_{st}) \chi^{(3)}(\omega_{pu} - \omega_{st}) \\
 &\quad \times \eta^*(\omega'_{pu}) \eta(\omega'_{st}) \eta^*(\omega'_{pr}) \eta(\omega_{pu}) \eta^*(\omega_{st}) \eta(\omega_{pu}) \\
 &\quad \times \langle 0 | \hat{a}^\dagger(\omega'_{pu}) \hat{a}(\omega'_{st}) \hat{a}^\dagger(\omega'_{pr}) \hat{a}(\omega_{pr}) \hat{a}^\dagger(\omega_{st}) \hat{a}(\omega_{pu}) | 0 \rangle \\
 &\quad \times \delta(\omega'_{pu} - \omega'_{st} + \omega'_{pr} - \omega_C) \delta(\omega_{pu} - \omega_{st} + \omega_{pr} - \omega_C), \tag{2}
 \end{aligned}$$

where $|0\rangle$ are the vacuum fields and ω' represents the frequency associated with the bra state.

The correlations between the field modes are caused by the bracketed part of the integrand in Eq. (2), which can be expanded as

$$\begin{aligned}
 &\langle 0 | \hat{a}^\dagger(\omega'_{pu}) \hat{a}(\omega'_{st}) \hat{a}^\dagger(\omega'_{pr}) \hat{a}(\omega_{pr}) \hat{a}^\dagger(\omega_{st}) \hat{a}(\omega_{pu}) | 0 \rangle \\
 &= v^*(\omega'_{pu}) \mu(\omega'_{st}) v^*(\omega'_{pr}) v(\omega_{pr}) \\
 &\quad \times \mu^*(\omega_{st}) v(\omega_{pu}) (D_{si} + D_{const}), \tag{3}
 \end{aligned}$$

where

$$\begin{aligned}
 D_{si} &= \delta(\omega'_{st} - 2\Omega + \omega_{pu}) \delta(\omega'_{pu} - 2\Omega + \omega_{st}) \delta(\omega'_{pr} - \omega_{pr}) \\
 &\quad + \delta(\omega'_{st} - 2\Omega + \omega_{pr}) \delta(\omega'_{pr} - 2\Omega + \omega_{st}) \delta(\omega'_{pu} - \omega_{pu}) \\
 &\quad + \delta(\omega'_{st} - 2\Omega + \omega_{pr}) \delta(\omega'_{pu} - 2\Omega + \omega_{st}) \delta(\omega'_{pr} - \omega_{pu}) \\
 &\quad + \delta(\omega'_{st} - 2\Omega + \omega_{pu}) \delta(\omega'_{pr} - 2\Omega + \omega_{st}) \delta(\omega'_{pu} - \omega_{pr}) \tag{4}
 \end{aligned}$$

and

$$\begin{aligned}
 D_{const} &= \delta(\omega'_{pu} - \omega_{pu}) \delta(\omega'_{st} - \omega_{st}) \delta(\omega'_{pr} - \omega_{pr}) \\
 &\quad + \delta(\omega'_{pu} - \omega_{pr}) \delta(\omega'_{st} - \omega_{st}) \delta(\omega'_{pr} - \omega_{pu}). \tag{5}
 \end{aligned}$$

D_{si} creates the correlation between the pump or probe (signal) field and the Stokes (idler) field. In contrast, the pump or probe (signal) field and the Stokes (idler) field are not correlated in the case of D_{const} .

When the condition of D_{si} is imposed, the six integral variables related to ω in Eq. (2) are reduced to three variables. In the following, we define the modes that are sensitive to the phase dispersion as the ω_l and ω'_l modes and the

insensitive modes as ω_m modes. For example, in the first term on the right-hand side of Eq. (4), $\delta(\omega'_{st} - 2\Omega + \omega_{pu})$ and $\delta(\omega'_{pu} - 2\Omega + \omega_{st})$ make the CARS spectrum sensitive to $H(\omega) = \eta(\omega)\eta(2\Omega - \omega)$; thus the ω_{pu} and ω'_{pu} modes in the first term are replaced with the ω_l and ω'_l modes, respectively. In contrast, $\delta(\omega'_{pr} - \omega_{pr})$ cancels out the effect of the phase dispersion of the relevant mode; thus it corresponds to the ω_m mode. The CARS spectrum enhanced with D_{si} is expressed as follows:

$$\begin{aligned}
 S_{si}(\omega_C, \tau) &= \int_{\Omega}^{2\Omega} d\omega_l \int_{\Omega}^{2\Omega} d\omega'_l \int_{\Omega}^{2\Omega} d\omega_m e^{i(\omega'_l - \omega_l)\tau} \\
 &\quad \times X(\omega_l, \omega'_l, \omega_m) H(\omega_l) M(\omega_l) H^*(\omega'_l) M(\omega'_l) M(\omega_m) \\
 &\quad \times \delta(\omega_l - 2\Omega + \omega'_l + \omega_m - \omega_C), \tag{6}
 \end{aligned}$$

where we use the approximation of $\mu(\omega) \simeq v(\omega)$ in the high-gain regime [14,15] and $M(\omega) = \mu(\omega)^2 = \mu(2\Omega - \omega)^2$ corresponds to the power spectrum of the signal or idler beam. The material response function is as follows:

$$\begin{aligned}
 X(\omega_l, \omega'_l, \omega_m) &= |\chi^{(3)}(\omega_l - 2\Omega + \omega'_l)|^2 \\
 &\quad + \chi^{(3)*}(\omega_l - 2\Omega + \omega_m) \chi^{(3)}(\omega'_l - 2\Omega + \omega_m) \\
 &\quad + \chi^{(3)*}(\omega_l - 2\Omega + \omega'_l) \chi^{(3)}(\omega'_l - 2\Omega + \omega_m) \\
 &\quad + \chi^{(3)*}(\omega_l - 2\Omega + \omega_m) \chi^{(3)}(\omega'_l - 2\Omega + \omega_l). \tag{7}
 \end{aligned}$$

The CARS spectrum produced with D_{const} is expressed as follows:

$$\begin{aligned}
 S_{\text{const}}(\omega_C) &= \int_{\Omega}^{2\Omega} d\omega_{\text{pr}} \int_0^{\Omega} d\omega_{\text{st}} \int_{\Omega}^{2\Omega} d\omega_{\text{pu}} [|\chi^{(3)}(\omega_{\text{pu}} - \omega_{\text{st}})|^2 \\
 &\quad + \chi^{(3)*}(\omega_{\text{pr}} - \omega_{\text{st}}) \chi^{(3)}(\omega_{\text{pu}} - \omega_{\text{st}})] \\
 &\quad \times M(\omega_{\text{pr}}) M(\omega_{\text{st}}) M(\omega_{\text{pu}}) \\
 &\quad \times \delta(\omega_{\text{pu}} - \omega_{\text{st}} + \omega_{\text{pr}} - \omega_C). \quad (8)
 \end{aligned}$$

This equation does not include $H(\omega)$, unlike Eq. (6), and thus it is not affected by phase dispersion. From these calculations, it can be seen that the total spectrum of CARS driven by twin beams can be expressed as the sum of the τ -dependent part [$S_{\text{si}}(\omega_C, \tau)$] and the τ -independent part [$S_{\text{const}}(\omega_C)$].

C. Delay-dependent CARS light intensity and temporal-correlation profile

We provide the light intensity of CARS driven by twin beams. The CARS light intensity is proportional to $G(\tau) = G_{\text{si}}(\tau) + G_{\text{const}}$ [13]. The terms on the right-hand side are, respectively, expressed as

$$\begin{aligned}
 G_{\text{si}}(\tau) &= \frac{1}{8\pi^3} \int_{\Omega}^{2\Omega} d\omega_l \int_{\Omega}^{2\Omega} d\omega'_l \int_{\Omega}^{2\Omega} d\omega_m e^{i(\omega'_l - \omega_l)\tau} \\
 &\quad \times H(\omega_l) M(\omega_l) H^*(\omega'_l) M(\omega'_l) \\
 &\quad \times M(\omega_m) X(\omega_l, \omega'_l, \omega_m) \quad (9)
 \end{aligned}$$

and

$$\begin{aligned}
 G_{\text{const}} &= \frac{1}{8\pi^3} \int_{\Omega}^{2\Omega} d\omega_{\text{pr}} \int_0^{\Omega} d\omega_{\text{st}} \int_{\Omega}^{2\Omega} d\omega_{\text{pu}} \\
 &\quad \times [|\chi^{(3)}(\omega_{\text{pu}} - \omega_{\text{st}})|^2 + \chi^{(3)*}(\omega_{\text{pr}} - \omega_{\text{st}}) \\
 &\quad \times \chi^{(3)}(\omega_{\text{pu}} - \omega_{\text{st}})] M(\omega_{\text{pr}}) M(\omega_{\text{st}}) M(\omega_{\text{pu}}), \quad (10)
 \end{aligned}$$

where the spectra of $G_{\text{si}}(\tau)$ and G_{const} correspond to $S_{\text{si}}(\omega_C, \tau)$ and $S_{\text{const}}(\omega_C)$, respectively.

Here, we show that the shape of the temporal-correlation profile can be determined by measuring the τ -dependent NCARS intensity. Under the assumption of an NCARS process, namely, $\chi^{(3)} = \chi_{\text{NR}}^{(3)}$, Eqs. (9) and (10) can be rewritten, respectively, as follows:

$$\begin{aligned}
 G_{\text{si, w/o}}(\tau) &= \frac{|\chi_{\text{NR}}^{(3)}|^2}{2\pi^3} \left| \int_{\Omega}^{2\Omega} d\omega_l H(\omega_l) M(\omega_l) e^{-i\omega_l \tau} \right|^2 \\
 &\quad \times \int_{\Omega}^{2\Omega} d\omega_m M(\omega_m), \quad (11)
 \end{aligned}$$

$$G_{\text{const, w/o}} = \frac{|\chi_{\text{NR}}^{(3)}|^2}{4\pi^3} \left[\int_{\Omega}^{2\Omega} d\omega M(\omega) \right]^2, \quad (12)$$

where the subscript w/o means that $\chi^{(3)} = \chi_{\text{NR}}^{(3)}$ is implied. The function of $\left| \int_{\Omega}^{2\Omega} d\omega_l H(\omega_l) M(\omega_l) e^{-i\omega_l \tau} \right|^2$ in Eq. (11) corresponds to the temporal-correlation profile of the twin beams. The temporal-correlation profile can also be measured using other nonlinear optical frequency conversions such as the sum-frequency generation process [1–4].

D. Method for extracting effect of twin-beam correlation

Here, we explain how to extract $G_{\text{si}}(\tau)$ and $S_{\text{si}}(\omega_C, \tau)$ from the CARS intensity and spectrum. Although the total CARS intensity [$G(\tau)$] includes the effects of both $G_{\text{si}}(\tau)$ and G_{const} , G_{si} can be made to approach 0 by making τ sufficiently larger than the temporal correlation width of the twin beams and the reciprocal of $\Gamma_{\text{Ram},j}$, namely, $G(\tau \rightarrow \infty) = G_{\text{const}}$. The CARS light intensity $G_{\text{si}}(\tau)$, in which we are interested, can be extracted as follows:

$$G_{\text{si}}(\tau) = G(\tau) - G(\tau \rightarrow \infty). \quad (13)$$

The same relationship also holds for the CARS spectra:

$$S_{\text{si}}(\omega_C, \tau) = S(\omega_C, \tau) - S(\omega_C, \tau \rightarrow \infty). \quad (14)$$

E. Spectral focusing by twin beams: Effect of second-order dispersion on correlation-enhanced CARS spectra

We now investigate the effect of the phase dispersion on the spectral resolution of CARS spectroscopy. As a preparation step, we first perform variable conversion for Eq. (6):

$$\omega_l = \frac{\omega_L}{2} + \frac{\omega_C - \omega_m}{2} + \Omega, \quad (15)$$

$$\omega'_l = -\frac{\omega_L}{2} + \frac{\omega_C - \omega_m}{2} + \Omega, \quad (16)$$

$$\omega_m = \Omega_{\text{sig}} + \omega_{\delta m}, \quad (17)$$

where Ω_{sig} is the peak angular frequency of the signal beam. We then obtain the following equations:

$$\begin{aligned}
S_{\text{si}}(\omega_C, \tau) = & \frac{1}{4} \int_{\Omega_{\text{id}} - \Omega}^{\Omega_{\text{id}}} d\omega_{\delta m} \int_{-[\omega_C - (\omega_{\delta m} + \Omega_{\text{sig}})]}^{\omega_C - (\omega_{\delta m} + \Omega_{\text{sig}})} d\omega_L e^{-i\omega_L \tau} \\
& \times H\left(\frac{\omega_L}{2} + \frac{\omega_C - \omega_{\delta m} + \Omega_{\text{id}}}{2}\right) H^*\left(-\frac{\omega_L}{2} + \frac{\omega_C - \omega_{\delta m} + \Omega_{\text{id}}}{2}\right) \\
& \times M\left(\frac{\omega_L}{2} + \frac{\omega_C - \omega_{\delta m} + \Omega_{\text{id}}}{2}\right) M\left(-\frac{\omega_L}{2} + \frac{\omega_C - \omega_{\delta m} + \Omega_{\text{id}}}{2}\right) \\
& \times M(\Omega_{\text{sig}} + \omega_{\delta m}) X'(\omega_L, \omega_C, \omega_{\delta m}), \tag{18}
\end{aligned}$$

where Ω_{id} is the peak angular frequency of the idler beam. The material response function is expressed as

$$\begin{aligned}
X'(\omega_L, \omega_C, \omega_{\delta m}) = & X(\omega_L, \omega'_L, \omega_m) \\
= & |\chi^{(3)}(\omega_C - \Omega_{\text{sig}} - \omega_{\delta m})|^2 + \chi^{(3)*} \left(\frac{\omega_L}{2} + \frac{\omega_C + \omega_{\delta m} - \Omega_{\text{id}}}{2} \right) \\
& \times \chi^{(3)} \left(-\frac{\omega_L}{2} + \frac{\omega_C + \omega_{\delta m} - \Omega_{\text{id}}}{2} \right) \\
& + \chi^{(3)*}(\omega_C - \Omega_{\text{sig}} - \omega_{\delta m}) \chi^{(3)} \left(-\frac{\omega_L}{2} + \frac{\omega_C + \omega_{\delta m} - \Omega_{\text{id}}}{2} \right) \\
& + \chi^{(3)*} \left(\frac{\omega_L}{2} + \frac{\omega_C + \omega_{\delta m} - \Omega_{\text{id}}}{2} \right) \chi^{(3)}(\omega_C - \Omega_{\text{sig}} - \omega_{\delta m}). \tag{19}
\end{aligned}$$

For simplicity, we consider only the second-order phase factor, namely $H(\omega) = \exp[-iD_2(\omega - \Omega_{\text{sig}})^2]$. $M(\omega)$ is assumed to have a Gaussian shape, $M(\omega) = \exp[-(\omega - \Omega_{\text{sig}})^2/2\sigma^2]$. With these substitutions, the CARS spectrum enhanced with D_{si} [Eq. (18)] becomes

$$\begin{aligned}
S_{\text{si}}(\omega_C, \tau) = & \frac{1}{4} \int_{\Omega_{\text{id}} - \Omega}^{\Omega_{\text{id}}} d\omega_{\delta m} e^{-\frac{\omega_{\delta m}^2}{2\sigma^2}} e^{-\frac{(\omega_C - \Omega_{\text{NC}} - \omega_{\delta m})^2}{4\sigma^2}} \\
& \times \int_{-[\omega_C - (\omega_{\delta m} + \Omega_{\text{sig}})]}^{\omega_C - (\omega_{\delta m} + \Omega_{\text{sig}})} d\omega_L g(\omega_L, \omega_C, \omega_{\delta m}) \\
& \times X'(\omega_L, \omega_C, \omega_{\delta m}) e^{-i\omega_L \tau}, \tag{20}
\end{aligned}$$

where $\Omega_{\text{NC}} = 2\Omega_{\text{sig}} - \Omega_{\text{id}}$ and $g(\omega_L, \omega_C, \omega_{\delta m}) = e^{-iD_2(\omega_C - \Omega_{\text{NC}} - \omega_{\delta m})\omega_L} e^{-(\omega_L^2/4\sigma^2)}$. The integral with respect to ω_L in Eq. (20) is approximately proportional to $\tilde{g}X'$, which represents the Fourier transformation of the function gX' from the ω_L domain to the τ domain. Equation (20) is expressed using the convolution theorem as follows:

$$\begin{aligned}
S_{\text{si}}(\omega_C, \tau) \propto & \int_{\Omega_{\text{id}} - \Omega}^{\Omega_{\text{id}}} d\omega_{\delta m} e^{-\frac{\omega_{\delta m}^2}{2\sigma^2}} \\
& \times \int_{-\infty}^{\infty} d\tau' G(\omega_C, \omega_{\delta m}, \tau - \tau') \tilde{X}'(\omega_C, \omega_{\delta m}, \tau'), \tag{21}
\end{aligned}$$

where

$$G(\omega_C, \omega_{\delta m}, \tau) = e^{-\frac{(\omega_C - \omega_{\delta m} - \Omega_{\text{NC}})^2}{4\sigma^2}} e^{-\sigma^2[\tau + D_2(\omega_C - \omega_{\delta m} - \Omega_{\text{NC}})]^2}. \tag{22}$$

The functions contained in Eq. (21) have the following physical meanings. The function $\tilde{X}'$ is the Fourier transform of the material response function that we want to measure. The function $G(\omega_C, \omega_{\delta m}, \tau)$ determines the wave-number range of CARS driven by the twin beam. The Gaussian function $\exp(-\omega_{\delta m}^2/2\sigma^2)$ in Eq. (21) is convolved with $G(\omega_C, \omega_{\delta m}, \tau)\tilde{X}'$. This convolution reduces the wave-number resolution of CARS spectroscopy, therefore the effect of $\omega_{\delta m}$ is discussed below. On the other hand, the integration with respect to τ' has the effect of reducing the resolution in the temporal domain and this effect is not the focus of this study.

The effect of narrowing the instantaneous spectral width due to second-order dispersion is exploited in conventional spectral focusing using ultrashort pulses. It can be seen from Eq. (21) that a similar effect also occurs in the case of twin beams. In Fig. 3(a), we show the spectral range of CARS driven by twin beams when $\omega_{\delta m} = 0$, [i.e., the full width at half maximum (FWHM) range for $G(\omega_C, 0, \tau)$]. In the figure, the cases in which $D_2 = 0$ and $D_2 \neq 0$ are represented by solid ellipses with transparent and blue inner regions, respectively. When D_2 takes a nonzero value, the peak position for each ω_C value shifts along the τ direction depending on the ω_C value; i.e., frequency ω_C exhibits

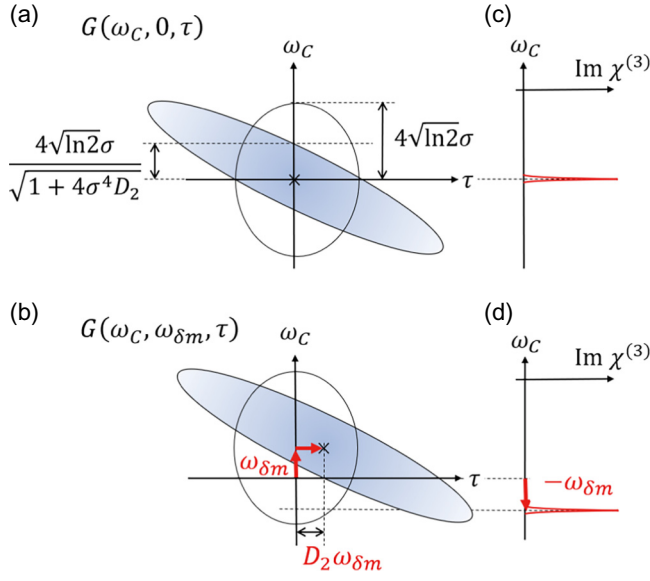


FIG. 3. $G(\omega_C, \omega_{\delta m}, \tau)$ and the imaginary part of the complex Lorentz function $[\text{Im } \chi^{(3)}]$ with $\Omega_{\text{Ram}} = \Omega_{\text{sig}} - \Omega_{\text{id}}$. (a) $G(\omega_C, 0, \tau)$. (b) $\text{Im } \chi$ for $\omega_{\delta m} = 0$. (c) $G(\omega_C, \omega_{\delta m}, \tau)$. (d) $\text{Im } \chi$ for $\omega_{\delta m} \neq 0$.

chirping. This causes the narrowing of the instantaneous frequency width of ω_C for a fixed τ , similar to the case for a chirped ultrashort pulse [13]. For example, the FWHM value decreases from $4\sqrt{\ln 2}\sigma$ to $4\sqrt{\ln 2}\sigma/\sqrt{1+4\sigma^2 D_2}$. When $\omega_{\delta m}$ becomes nonzero, as indicated by the red arrows in Fig. 3(b), the position is shifted by $\omega_{\delta m}$ in the direction of ω_C and by $D_2\omega_{\delta m}$ in the direction of τ from $G(\omega_C, 0, \tau)$.

From Eq. (21), it can be seen that the deterioration in wave-number resolution is caused by the superimposition of the spectra with $\omega_{\delta m} \neq 0$ on the spectrum with $\omega_{\delta m} = 0$. Therefore, to show the improvement in wave-number resolution due to the narrowing of the instantaneous frequency width, it is necessary to show that the contribution of the spectra with $\omega_{\delta m} \neq 0$ is reduced. As a simple example, we consider CARS spectroscopy for a Raman spectrum centered at a wave number of $\Omega_{\text{Ram}} = \Omega_{\text{sig}} - \Omega_{\text{id}}$. In Fig. 3(c), we show the corresponding imaginary part of the complex Lorentz function ($\text{Im } \chi^{(3)}$) with $\omega_{\delta m} = 0$, in which the Raman peak matches the center of $G(\omega_C, 0, \tau)$, and the largest signal is obtained when $\tau = 0$. When $\omega_{\delta m}$ is not zero, the function $\chi^{(3)}$ shifts on the ω_C axis depending on the variables of $\chi^{(3)}$ in Eq. (19). The most important terms that cause an improvement in the wave-number resolution are $\chi^{(3)*}\{(\omega_L/2) + [(\omega_C + \omega_{\delta m} - \Omega_{\text{id}})/2]\}$ and $\chi^{(3)}\{-(\omega_L/2) + [(\omega_C + \omega_{\delta m} - \Omega_{\text{id}})/2]\}$. In these terms, because the variable includes $\omega_C + \omega_{\delta m}$, it shifts in the opposite direction to $G(\omega_C, \omega_{\delta m}, \tau)$, as shown in Fig. 3(d). As a result, as D_2 increases, the value produced through multiplication with $G(\omega_C, \omega_{\delta m}, \tau)$ at $\tau = 0$ decreases, so the contribution of the spectra with $\omega_{\delta m} \neq 0$ is reduced.

III. EXPERIMENTAL APPARATUS

A. Outline of experimental setup

In the experiment, the fundamental light source was a Q-switched laser that emitted light pulses at a wavelength of 1064 nm with a FWHM value of 3.9 ns and a repetition rate of 7.0 kHz (Cobolt, Tor 1064-nm laser). The fundamental beam was frequency doubled by type-I second-harmonic generation in a 15-mm lithium triborate (LBO) crystal (Edmund Optics, #11-173). The LBO crystal was housed in a custom-made oven and temperature-stabilized by a proportional-integral-derivative-controlled Peltier element (Thorlabs, TED200C). The second harmonic at $\lambda_{2\Omega} = 532.2$ nm with an FWHM value of 3.5 ns was used as the excitation source for PDC. The wavelength FWHM value of the second harmonic was narrower than 0.1 nm. As shown in Fig. 1, the intense signal and idler twin beams were generated by nanosecond-pumped single-pass PDC in a temperature-controlled periodically poled MgO:LiNbO₃ (PPLN) crystal. The PDC conversion efficiency was about 0.3% for 500 mW of excitation mean power. The twin beams from the PPLN were collimated by a lens (focal length 35 mm). They were separated by a dichroic mirror (Edmund Optics, #87-041). The delay, τ , of the idler beam relative to the signal beam was then tuned. After the twin beams were combined, they were focused on the Raman sample by an objective lens (Mitutoyo Plan Apo NIR B 20 $\times$, numerical aperture 0.4). The focusing relaxes the phase-matching condition [16,17]. The intensity and spectra of CARS light from the sample were detected by an electron-multiplying charge-coupled device (EMCCD; Princeton Instruments, ProEM-HS 512BX3-G) and a spectrometer (Andor Technology, SR-500i-A and U416A-LDC-DD), respectively.

B. Twin-beam spectra and Raman sample

We evaluate the tunability of the wave-number difference of the signal and idler beams by measuring the peak wavelength of the signal beam (λ_{sig}) at various PPLN temperatures [a typical spectrum is shown in Fig. 4(a)]. In Fig. 4(b), we show the values of $(1/\lambda_{\text{sig}}) - (1/\lambda_{\text{id}})$ as a function of the PPLN temperature, where the peak wavelength of the idler beam is estimated as $\lambda_{\text{id}} = [(1/\lambda_{2\Omega}) - (1/\lambda_{\text{sig}})]^{-1}$. This result shows that our twin-beam source can be tuned from 1700 cm^{-1} to 3200 cm^{-1} . The lower limit of the wave number is due to the cutoff wavelength of the dichroic mirror in our experiment.

The sample was a 0.01-mm-thick polyethylene film (purchased from a local supermarket). In Fig. 5, we show the Raman spectrum measured by spontaneous Raman-scattering spectroscopy. In the CARS-spectroscopy experiment, two large peaks at 2845 cm^{-1} and 2880 cm^{-1} , respectively, separated by 35 cm^{-1} , were used for the experimental evaluation of the wave-number resolution.

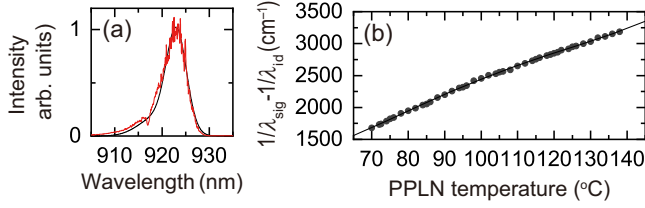


FIG. 4. Evaluation of the tunability of the wave-number difference of the twin beams. (a) A typical example of a signal spectrum at a PPLN temperature of 121.5°C. The red curve shows the experimental data. The black curve shows the spectral shape estimated from the spectrum of the NCARS light measured at $\tau = 4$ ps. (b) The wave-number difference for the twin beams as a function of the PPLN temperature. λ_{sig} and λ_{id} represent the peak wavelengths of the signal and idler beams, respectively.

IV. EXPERIMENTAL AND NUMERICAL RESULTS AND DISCUSSION

A. Characterization of twin-beam correlation by NCARS process

We measured the temporal-correlation profile for the twin beams using a 0.17-mm-thick glass plate with no Raman transitions in the region of interest. Note that the enhanced NCARS intensity from the glass plate is expressed by Eq. (11). Thus, the τ -dependent shape of the intensity corresponds to the twin-beam temporal-correlation profile. In Figs. 6(a)–6(c), we show the NCARS light intensity as a function of τ , where the vertical axes are normalized using the NCARS intensity measured around ± 4 ps. In these data, the temperature of the PPLN was set such that the wavelength difference for the twin beams was in resonance or near resonance with the Raman transitions for polyethylene [117°C, 119°C, and 121.5°C for Fig. 6(a), Fig. 6(b), and Fig. 6(c), respectively]. The ranges over which the intensity was enhanced (greater than 1) are about -0.7 to 1.0 ps in Figs. 6(a) and 6(b) and about -0.2 to 1.4 ps in Fig. 6(c).

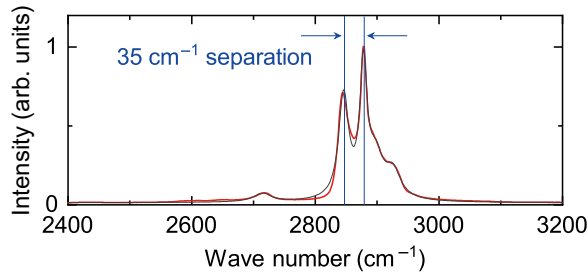


FIG. 5. Characterization of the Raman bands of polyethylene measured by spontaneous Raman spectroscopy. The red and black curves represent the experimental and fitting results, respectively. The two large peaks separated by 35 cm^{-1} were used to experimentally evaluate the wave-number resolution of the CARS spectroscopy.

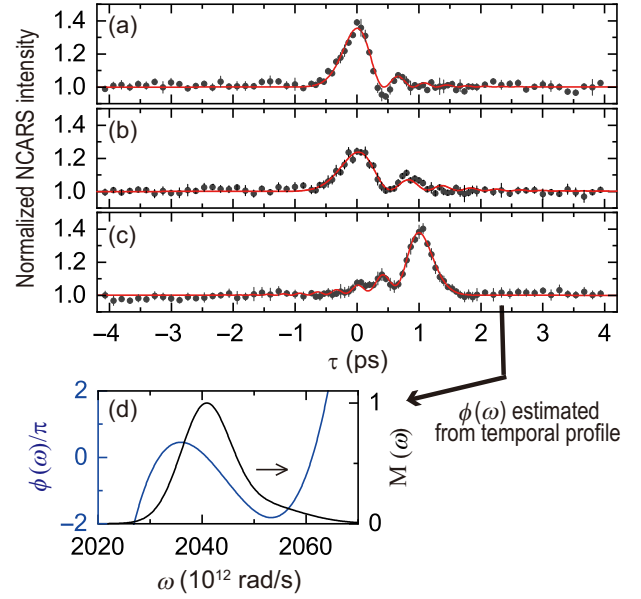


FIG. 6. Temporal-correlation profiles for twin beams and the estimated accumulation phase: the τ dependence of the NCARS intensity measured using glass for PPLN temperatures of (a) 117°C, (b) 119°C, and (c) 121.5°C. The black circles and red curves represent the experimental and fitting results, respectively. The vertical axes are normalized using the NCARS intensity measured around ± 4 ps. (d) ϕ estimated from the temporal-correlation profile measured in (c). $M(\omega)^2$ was estimated such that the spectrum of NCARS light for $\tau = 4$ ps is reproduced.

In Figs. 7(a)–7(c), we show the experimentally observed spectra for the enhanced NCARS light. These data were obtained by measuring the NCARS spectrum as a function

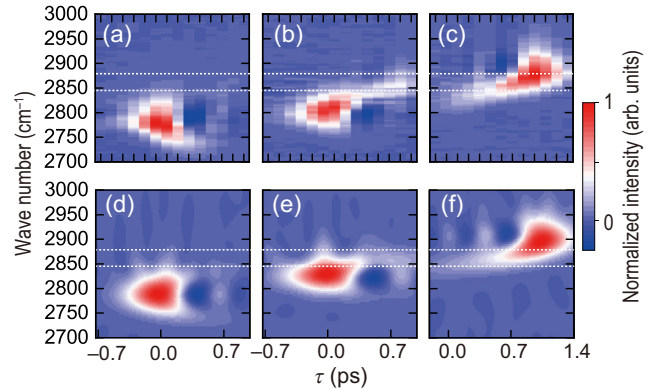


FIG. 7. Contour plots of the τ -dependent spectra for enhanced NCARS light. The top and bottom panels, respectively, show experimental and numerical results obtained for PPLN temperatures of (a),(d) 117°C, (b),(e) 119°C, and (c),(f) 121.5°C. These spectra were obtained by subtracting the spectrum measured around $\tau = \pm 4$ ps. The vertical axis shows the wave number, which was calculated as $k = \frac{2}{3}[(1/\lambda) - (1/2\lambda_2\Omega)]$ from the wavelength of NCARS light λ .

of τ and then subtracting the mean spectrum measured around $\tau = \pm 4$ ps. The vertical axis shows the wave number (k), which was calculated from the wavelength of NCARS light (λ). Since the signal and idler peak frequencies are the same amount $\delta\Omega$ away from Ω in the opposite direction, the corresponding λ and k^{-1} are proportional to $(3\delta\Omega + \Omega)^{-1}$ and $(2\delta\Omega)^{-1}$, respectively. From these relations, $k = \frac{2}{3}[(1/\lambda) - (1/2\lambda_{2\Omega})]$. A comparison of the overlap between the contour plots for NCARS and the two large Raman peaks for polyethylene (the two white dotted lines in Fig. 7) indicates that the two Raman peaks can be driven when the PPLN temperature is set to 121.5 °C [Fig. 7(c)]. In addition, in Fig. 7(c), the peak wave number of the NCARS light increases with increasing τ . This shift indicates that frequency chirping occurs due to the second-order dispersion shown in Fig. 3(a). Spectral focusing is expected to be feasible in this condition.

We performed a numerical simulation using Eq. (6) to provide support that the frequency chirping observed in Fig. 7(c) is caused by phase dispersion. As a first step for the simulation, we estimated $M(\omega)$. $M(\omega)$ is expected to match the spectrum of the signal beam (or idler beam) if the phase matching conditions are perfectly satisfied in the NCARS process. However, when the phase-matching conditions are only partially satisfied, $M(\omega)$ will deviate from the signal spectrum. To account for the effect of partial phase mismatch in the NCARS process, we did not directly use the signal spectrum shown by the red curve in Fig. 4(a) as $M(\omega)$. Instead, we estimated $M(\omega)$ such that it reproduces the spectrum of the NCARS light measured at $\tau = 4$ ps. The estimate for 121.5 °C is shown as the black curve in Fig. 4(a) [the same result is shown in Fig. 6(d)]. The shape of the signal spectrum [red curve in Fig. 4(a)] is in good agreement with the estimate, implying that phase matching is well satisfied in the NCARS process.

$H(\omega)$ was estimated from a fitting to the temporal-correlation profiles using Eq. (11). In this fitting, we assumed up to third-order dispersion as follows:

$$H(\omega) = e^{-2i\phi(\omega)} = e^{-2i[D_1(\omega - \Omega_{\text{sig}}) + \frac{D_2}{2!}(\omega - \Omega_{\text{sig}})^2 + \frac{D_3}{3!}(\omega - \Omega_{\text{sig}})^3]}. \quad (23)$$

The fitting results are shown by the red curves in Figs. 6(a)–6(c). As a representative example, $\phi(\omega)$ obtained from the fitting for 121.5 °C is shown in Fig. 6(d).

In Figs. 7(d)–7(f), we show the simulation results calculated by substituting the estimated $M(\omega)$ and $H(\omega)$ into Eq. (6) with $X(\omega_l, \omega_i, \omega_m) = \text{const.}$ The good agreement between the experiment and simulation means that the complex shapes of the spectrum are caused by the accumulated phase by the optical elements and spatial diffraction. This agreement is strong evidence that the frequency chirping observed in Fig. 7(c) is caused by second-order phase dispersion.

B. CARS spectroscopy with no correlation between twin beams

We performed CARS spectroscopy with no correlation between the twin beams by setting $\tau = 4$ ps. This spectroscopy corresponds to the observation of the CARS spectrum with D_{const} [Eq. (8)]. In Fig. 8(a), we show a comparison between the CARS intensity from the polyethylene and the NCARS intensity from the glass as a function of the PPLN temperature, where the intensities are normalized by the intensity at 70 °C. The CARS intensity from the polyethylene exhibits a clear peak structure that originates from Raman transitions centered at 119 °C, corresponding to a wave number of 2834 cm^{-1} .

In Figs. 8(b) and 8(c), respectively, we show the CARS spectra and NCARS spectra obtained at 117 °C, 119 °C, and 121.5 °C, where each spectrum intensity is normalized by the peak value. For 117 °C and 119 °C, the peaks in the CARS spectrum are shifted to higher wave numbers compared to those of NCARS. For 121.5 °C, the wave-number peak of the CARS spectrum shifts to almost the center of the two Raman peaks. Although these peak shifts are thought to be caused by the two large Raman transitions, the two Raman peaks cannot be distinguished at any PPLN temperature. This indicates that the spectral resolution of CARS spectroscopy with uncorrelated twin beams is worse than 35 cm^{-1} . This result is consistent with the wave-number resolution of 42 cm^{-1} , which is estimated

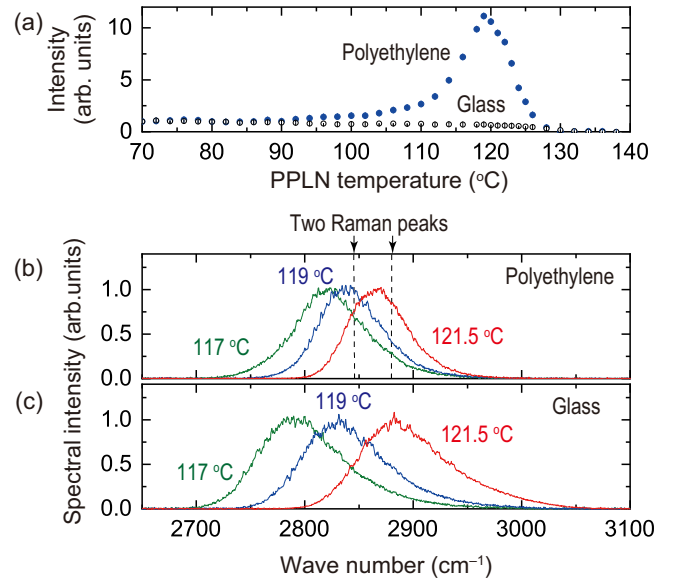


FIG. 8. CARS spectroscopy without twin-beam correlation. (a) A comparison between the CARS intensity of polyethylene and the NCARS intensity of glass as function of the PPLN temperature. (b) CARS spectra measured at PPLN temperatures of 117 °C, 119 °C, and 121.5 °C. (c) NCARS spectra measured at these temperatures. The spectral intensity is normalized by the maximum value of each spectrum.

from the FWHM of the spectrum of the signal beam for a PPLN temperature of 121.5°.

C. CARS spectroscopy with twin-beam correlation

We performed CARS spectroscopy with twin-beam correlation, which corresponds to the observation of the CARS spectrum with D_{si} [Eq. (6)]. In Figs. 9(a)–9(c), we show the experimentally observed spectra of enhanced CARS light, which were obtained by measuring the CARS spectrum as a function of τ and then subtracting the average spectrum measured around $\tau = \pm 4$ ps from the CARS spectrum. For a PPLN temperature of 117°C, the spectral shape [Fig. 9(a)] is similar to that of the enhanced NCARS light [Fig. 7(a)]. When the temperature was set to 121.5°C [Fig. 9(c)], the spectral shape became quite different from that of the enhanced NCARS light [Fig. 7(c)]. The difference between CARS and NCARS is thought to be caused by the two Raman peaks for polyethylene (two white dotted lines), since the spectral intensities of the enhanced CARS are concentrated at the Raman resonance peaks.

To investigate the effect of spectral focusing, we focus on the results for a PPLN temperature of 121.5°C. It should be noted that at this temperature, the wave-number shift (frequency chirping) occurs over the 35-cm⁻¹-separated Raman peaks as shown in Fig. 7(c). In Fig. 10(a), we compare the spectra of the enhanced CARS light from polyethylene and the NCARS light from glass for τ from -200 fs to 330 fs, which correspond to the partial results in Figs. 9(c) and 7(c). It can be seen that double dispersive profiles occur only in the enhanced CARS light around the two Raman peaks (black dotted lines). The dispersive profiles can be interpreted as resulting from the interference of resonant CARS and NCARS light [18]. The experimental observation of two dispersive profiles means

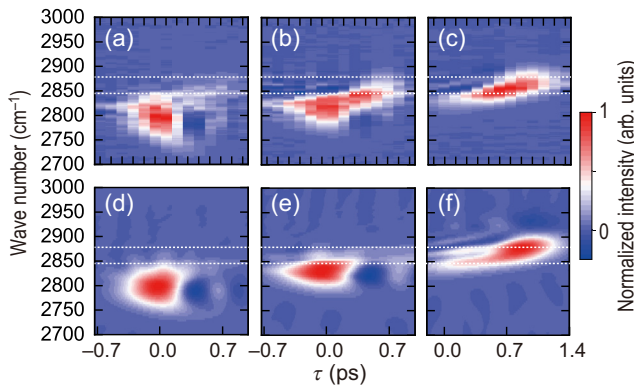


FIG. 9. Contour plots of the τ -dependent spectra for the enhanced CARS light. The top and bottom panels, respectively, show experimental and numerical results obtained for PPLN temperatures of (a),(d) 117°C, (b),(e) 119°C, and (c),(f) 121.5°C. These spectra were obtained by subtracting the spectrum measured around $\tau = \pm 4$ ps.

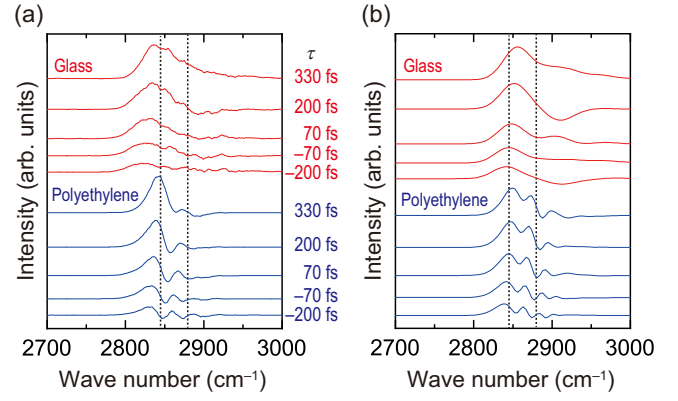


FIG. 10. A comparison of the spectra of enhanced CARS light from polyethylene with those of enhanced NCARS light from glass for τ below 330 fs: (a) experimental and (b) numerical results. (a) is the same as the partial result in Figs. 9(c) and 7(c). (b) is the same as the partial result in Figs. 9(f) and 7(f).

that the two Raman peaks for the polyethylene can be distinguished. In contrast, the two large Raman spectral peaks cannot be distinguished in the CARS spectrum with uncorrelated twin beams [Fig. 8(b)] and thus these results indicate that the spectral resolution is improved by the effect of twin-beam correlation. Since two Raman peaks can be distinguished, the improved spectral resolution is expected to be at least better than 35 cm⁻¹.

To confirm that the dispersive profiles observed in Fig. 10(a) are caused by phase dispersion, we first performed a numerical simulation using Eq. (6) for the case of $H(\omega) = 1$ (i.e., without phase dispersion). In Figs. 11(a) and 11(b), we show that the simulation results for glass and polyethylene for a PPLN temperature of 121.5°C. The spectral shape for glass [Fig. 11(a)] is different from the experimental result [Fig. 7(f)], showing a symmetrical shape with respect to positive and negative τ . Furthermore, as we expected, the numerical simulation for polyethylene shows that the dispersive structures do not appear in the case of $H(\omega) = 1$ [Fig. 11(b)].

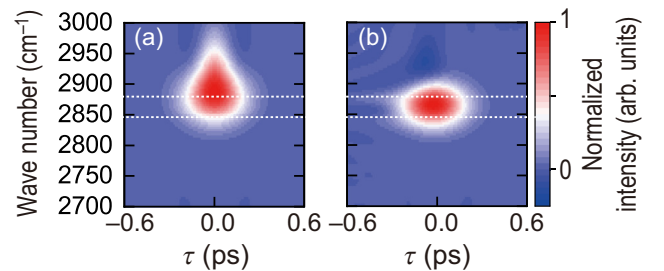


FIG. 11. Contour plots of the spectra of enhanced NCARS and CARS light without phase dispersion [$H(\omega) = 1$] as function of delay τ and wave number k . Results are shown for (a) glass and (b) polyethylene.

We numerically simulated the experimental results in Figs. 9(a)–9(c) using Eq. (6) with phase dispersion. In this simulation, $\chi_{\text{NR}}^{(3)}$ was determined to reproduce the results of Fig. 9(c). From the simulation results shown in Figs. 9(d)–9(f), the CARS intensity is enhanced near the Raman resonance, similar to the experimental results. Furthermore, it can be seen that at 121.5 °C, multidispersive structures appear, as shown in Fig. 10(b). The reproduction of these experimental results strongly suggests that the improvement in spectral resolution observed in the experiment is caused by frequency chirping due to phase dispersion. One difference between the experiment and the simulation is that the simulation predicts the formation of a clear additional dispersive shape at a wave number higher than two dispersive profiles. One possible cause of this difference is the deviation between the actual and estimated values for $H(\omega)$.

V. CONCLUSIONS

This study has presented both experimental and theoretical investigations of spectral-focusing CARS spectroscopy using bright correlated twin beams. The theoretical analysis has demonstrated that twin-beam correlations enable the realization of spectral focusing. Experimentally, we have achieved CARS spectroscopy using a twin-beam source excited by nanosecond pulses, which has demonstrated improved spectral resolution due to the frequency chirping of the twin beams. From a practical perspective, twin beams with nanosecond-pulsed excitation offer a compact and cost-effective light source system. Additionally, PDC provides the flexibility to tune the frequency difference over a wide range. Twin-beam CARS spectroscopy is considered a promising technology for reducing the technical barriers and costs associated with high-performance broadband CARS spectroscopy. This quantum technology holds significant potential to broaden the applications of CARS spectroscopy.

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DATA AVAILABILITY

The data that support the findings of this paper are not publicly available upon publication because it is not

technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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