Nonlinear spectroscopy in the single optical cycle regime

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The use of extremely short sub-5-fs pulses that became available recently provides obvious advantages to a spectroscopic experiment. Next to the very high temporal resolution, the broad bandwidth covering an impressive spectral window at once. However, the standard description applicable to multi-cycle pulses becomes questionable for the pulses that consist merely of a couple of optical fringes. The conventionally employed slowly varying envelope approximation, implying that the change of the pulse amplitude on the duration of an optical cycle is negligible compared to the magnitude of the amplitude itself, can no longer be maintained. Furthermore, the phase-matching bandwidth that is limited due to dispersion in the nonlinear medium rapidly gains importance with the broadening of the pulse spectrum. Another point of serious concern is the frequency-dependent variation in the sensitivity of the photodetector employed to register the signal generated in the nonlinear process. In combination, the above listed features of an experiment with broadband pulses result in what is known as a spectral filter effect. On top of that, artificial lengthening of the observed time dependences is a direct consequence of the nonlinear geometry explored in experiments.

In this contribution we present a theoretical analysis in which the frequency- and time-domain formalism of ultrafast nonlinear spectroscopy is thoroughly examined. The complete expressions valid even for single-cycle-pulse applications are derived for the nonlinear signal in the frequency and time domains. We also assert that the influence of geometrical delay smearing does not introduce a significant distortion of the observed traces provided that the geometry is carefully optimized. The derived formalism is applied to photon-echo spectroscopy on the hydrated electron with 5-fs pulses.

We consider the case of nonlinear geometry in which three beams intersect at a small angle in nonlinear medium. A typical equation that governs the propagation of the fourth, signal, field can be obtained directly from Maxwell's equations:

$$
\frac{\partial E_s(z,\Omega)}{\partial z} = -\frac{i\epsilon_0 c_0}{2\Omega} \int \int d\omega' d\omega'' E_r(z,\Omega') E_r(z,\Omega'') \delta(\Omega - \omega' + \omega'') 
$$

$$
\times \exp(\Delta k_s(\Omega,\omega',\omega'') z) - i\omega' \frac{1}{\omega' - \omega''} \int d\omega' d\omega'' \frac{d\phi(\omega',\omega'')}{\omega'}
$$

where $E_s$ stands for electric fields, $\chi^{(3)}$ is the third-order susceptibility, the phase mismatch is denoted as $\Delta k_s(\Omega,\omega',\omega'') = k_s(\omega') - k_s(\omega'') + k_s(\omega'') + k_s(\omega' + \omega'') - k_s(\omega' + \omega'')$, and $E_r(z,\Omega)$ is the resonant electric field.

Figure 2(a) and 2(b) present the two-pulse photon echo signals obtained from water and electrons solvated in water, respectively.
A minute difference in the widths of these two traces suggests that the electronic dephasing of the hydrated electrons is extremely fast. The finite population lifetime of the electrons in the excited state causes the delay of the echo trace in Fig. 2(b). The best fit to the experimental data yields the dephasing time of $T_2 = 1.6$ fs. With this value, we successfully modeled the absorption spectrum of the hydrated electrons by an extended Lorentzian line shape, which indicates the breakdown of the rotating wave approximation (RWA).


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**PMD probability distribution for arbitrary distances**

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Polarization mode dispersion (PMD) is caused by the random birefringence present in optical fibers. It can lead to pulse spreading and depolarization, and is detrimental to system performance. As transmission rates continue to increase, PMD has become a major impairment, thus motivating extensive experimental and theoretical study over the past few years.

PMD is characterized by a three-component dispersion vector $\mathbf{\Omega}$. Its magnitude $|\mathbf{\Omega}|$ gives the differential group delay (DGD) between the principle states, and its direction gives the orientation of the principle states of polarization on the Poincaré sphere at the output. For short distances, PMD is deterministic, and the DGD distribution is a $\delta$-function. For long distances, previous work assuming weak or completely randomizing birefringence models led to a Maxwellian DGD distribution. This long-distance result has been confirmed by numerous numerical and experimental studies. However, there has been as yet no analytic demonstration of this fact assuming a realistic model of the birefringence variation in the fiber. More importantly, there have been no careful studies of how long it takes to reach the asymptotic distribution. This may be important for calculating the penalties due to PMD in the transient regime, because the usual assumption that the distribution is Maxwellian may be erroneous.

In this work, we calculate the DGD distribution in the intermediate distance regime. We begin with the basic dynamical equation for dispersion vector $W$:

$$\frac{\partial W(z,\omega)}{\partial z} + \frac{\partial W(z,\omega)}{\partial \omega} + W(z,\omega) \times \mathbf{\Omega}(z,\omega).$$

where the vector $W$ represents the local birefringence in the fiber. We choose a simple realistic birefringence model (the first model of Wai and Menyuk), where the fiber is assumed to have linear birefringence of fixed strength $2b$, but where the birefringence orientation is assumed to be driven by a white noise process.

In other words, $W = (2b \cos \theta, 2b \sin \theta, 0)$, and $W(z,\omega) = \{E_z^0(\omega), E_{\varphi}^0(\omega)\}$, where $E_z^0(\omega)$ and $E_{\varphi}^0(\omega)$ are the electric field components along the $z$ and $\varphi$ axes, respectively.

For simplicity, we assume that $\mathbf{\Omega}$ is independent of $z$, and that the fiber is infinite in length. The differential group delay at various distances is

$$\frac{\partial P}{\partial Z} = 2 \left( \frac{\Omega_1^0}{\Omega_1^0 + \Omega_2^0} \right) \frac{\partial P}{\partial \Omega_2^0}. \tag{2}$$

Here $Z = z/b_{\text{fiber}}$, and $h_{\text{fiber}}$ is the typical fiber correlation length. The dotted lines in (b) and (c) are Maxwellian distributions for comparison.

$h_{\text{fiber}}$ is much larger than the beat length $\pi/b$.

In this case, the Fokker–Planck for $\mathbf{\Omega}$ can be averaged over the rapid birefringence rotation, and the probability density function $P$ for $\mathbf{\Omega}$ satisfies the following reduced equation:

$$\frac{\partial P}{\partial Z} = \frac{1}{2} \left( \frac{\Omega_1^0}{\Omega_1^0 + \Omega_2^0} \right) \frac{\partial P}{\partial \Omega_2^0}. \tag{2}$$

Here $Z = z/b_{\text{fiber}}$, and $b_{\text{fiber}}$ is approximately 0.32 ps. It is important to note, however, that Eq. (2) no longer has any free parameters. Thus, the