Nonlinear spectroscopy in the single optical cycle regime

M.S. Pshenichnikov, A. Ilduñka, D.A. Wiersma, Ultrashort Laser and Spectroscopy Lab., Univ. of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands E-mail: M.S.Pshenichnikov@chem.rug.nl

The use of extremely short sub-1 fs pulses that became available recently1,2 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 nicely of a couple of optical fringes. The conventionally employed slowly varying envelope approximation,1 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.3 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,4 on top of that, artificial lengthening of the observed time dependencies is a direct consequence of the noncollinear geometry employed 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 the two-pulse photon-echo spectroscopy on the hydrated electron with 5-fs pulses.

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

\[
\frac{\partial}{\partial z} E_s(z,\omega) = i \frac{c_0}{2\varepsilon_0} \int \int d\omega' d\omega'' \chi^{(3)}(\omega - \omega', \omega''; \omega - \omega' + \omega'') E_l(z,\omega') E_l(z,\omega''; \omega - \omega' + \omega'') \exp \left( i \Delta k_z (\omega, \omega', \omega'') z \right) d\omega' d\omega''
\]

where \(E_l\) stands for electric fields, \(\chi^{(3)}\) is the third-order susceptibility, the phase mismatch is denoted as \(\Delta k_z (\omega', \omega, \omega'') = -k_{12}(\omega') + k_{12}(\omega - \omega' + \omega'')\). For two pulses is suppressed.

Figures 2(a) and 2(b) present the two-pulse photon echo signals obtained from the 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).\(^1\)


---

CML 10:15 am

**Polarization Mode Dispersion**

Fred L. Heismann, Tyco Submarine Systems Ltd., USA, President

CML1 10:15 am

**Polarization mode dispersion in optical communication systems**

Curtis M. Menyuk, Univ. of Maryland–Baltimore Campus, 1000 Hilltop Cir., Baltimore MD 21250, USA

Polarization mode dispersion (PMD) has become one of the major barriers to achieving single-channel data rates of 10 Gbits/sec and beyond in optical communications systems. In this tutorial, the physical origins of birefringence and PMD are first reviewed. The effects of first-, second-, and higher-order PMD are next elucidated. Finally, PMD compensators and emulators are described. In particular, the need for physically realistic emulators in order to realistically evaluate the effects due to PMD and compensators is emphasized.

---

CML2 10:15 am

**PMD probability distribution for arbitrary distances**

Jianke Yang, William L. Kath,* Curtis M. Menyuk,** Department of Mathematics and Statistics, Univ. of Vermont, Burlington, Vermont 05401, USA; E-mail: jyang@uvm.edu

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 $\Omega$. Its magnitude $|\Omega|$ 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 has led to a Maxwellian DGD distribution.\(^2\) 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 $\Omega$:

$$\frac{d\Omega(z,\omega)}{dz} = \frac{g(\Omega(z,\omega))}{\omega} + W(z,\omega) \times \Omega(z,\omega),$$

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 $2\theta$, but where the birefringence orientation is assumed to be driven by a white noise process. In other words, $W = (2\theta \cos 2\theta \sin 0, 0, 0)$, and $dW/d\omega = g(\Omega)(s_{\text{local}}(z), \omega)$, where $g(\Omega)$ is $0$, and $g(\Omega)$ is the symmetric distribution in the (a, b) plane.

Here $Z = 1$ corresponds to one fiber correlation length, and the DGD unit is $1/\text{fiber correlation length}$. 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 equation for $\Omega$ can be averaged over the rapid birefringence rotation, and the probability density function $P$ for $\Omega$ satisfies the following reduced equation:\(^3\)

$$\frac{\partial P}{\partial Z} = \frac{1}{2} \left( \frac{\partial}{\partial \Omega_1} \Omega_2 \partial \Omega_3 \partial \Omega_1 \right) - \Omega_1 \frac{\partial^2 P}{\partial \Omega_1^2} \Omega_2 \Omega_3.$$

Here $Z = h_{\text{fiber}}$, and $\Omega_1 = \Omega_2 = \Omega_3 = 0$ for $h_{\text{fiber}}$. It is important to note, however, that Eq. (2) no longer has any free parameters. Thus, the...