Ultrashort-pulse measurement using noninstantaneous nonlinearities: Raman effects in frequency-resolved optical gating

Kenneth W. DeLong, Celso L. Ladera,* and Rick Trebino

MS 9057, Sandia National Laboratories, Livermore, California 94551-0969

Bern Kohler and Kent R. Wilson

Department of Chemistry, MS 0339, University of California, San Diego, La Jolla, California 92093-0339

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Ultrashort-pulse-characterization techniques generally require instantaneously responding media. We show that this is not the case for frequency-resolved optical gating (FROG). We include, as an example, the noninstantaneous Raman response of fused silica, which can cause errors in the retrieved pulse width of as much as 8% for a 25-fs pulse in polarization-gate FROG. We present a modified pulse-retrieval algorithm that deconvolves such slow effects and use it to retrieve pulses of any width. In experiments with 45-fs pulses this algorithm achieved better convergence and yielded a shorter pulse than previous FROG algorithms.

Ultrashort-pulse-characterization methods generally require nonlinear-optical media with essentially instantaneous response. For example, second-harmonic generation and multiphoton ionization are commonly used for autocorrelation measurements. Slower responses yield information equivalent to the pulse spectrum.[1] When full intensity-and-phase characterization is required, however, only nearly instantaneous processes can be used.[2,3] Unfortunately, these processes are often accompanied by noninstantaneous processes, as Raman ringing accompanies the electronic Kerr effect. In addition, nearly instantaneous effects tend to be weak, so use of a slower medium could extend the use of a technique to lower pulse energies. Thus it is important to develop measurement methods for use with slowly responding media.

In this Letter we show that frequency-resolved optical gating[3–5] (FROG), which involves generating a spectrogram of the pulse by using the pulse itself as a variable-delay gate, naturally accommodates media with noninstantaneous response. FROG uses an iterative pulse-retrieval algorithm and is in essence a deconvolution method. Here we consider the Raman effect, which necessarily accompanies the electronic Kerr effect in FROG, using the polarization-gate (PG) geometry. For fused silica it provides slight ringing on a ~30-fs time scale in the induced polarization and hence distorts experimental PG FROG traces. We show that, unaccounted for, it leads to retrieved pulses as much as 8% too long for the worst case of 25-fs pulses. We then present a modified algorithm, based on the method of generalized projections,[6] that accounts for the ringing—or, in principle, for any other noninstantaneous effect—and accurately retrieves the correct pulse in all cases. In an experimental trace obtained for a 45-fs pulse, the modified algorithm achieved lower rms error and a shorter pulse length than previous FROG algorithms, which assumed an instantaneous response.

The Raman response of fused silica for the case of self-action was studied by Stolen and co-workers,[7,8] who used experimentally measured Raman spectra. We use the formalism developed by Hellwarth,[9] which is valid in the Born–Oppenheimer approximation (where the optical frequencies are well below the electronic and well above the nuclear resonance frequencies), to write the nonlinear polarization in terms of response function integrals. Following Eqs. (4.9), (5.8), and (5.10) of Ref. 9, for the case of self-action in an isotropic medium we can write

$$P^{(3)}(t) = \frac{3}{2}\sigma E(t)|E(t)|^2 + 2E(t)\int_{-\infty}^{t} \mathrm{d}t'[a(t-t') + b(t-t')]|E(t')|^2, \tag{1}$$

where σ represents the strength of the fast nonlinearity and $a(t)$ and $b(t)$ are the isotropic and nonisotropic Raman contributions, respectively, of the nuclear motion to the nonlinear response. We ignore terms in the integral that oscillate on the order of twice the optical frequency because these terms vary quickly on the time scale of $a(t)$ and $b(t)$ (that of the nuclear motion) and therefore average to zero. The factor $N_{2\infty}$ (fast response) of Stolen *et al.*[7,8] is equal to $3\sigma/2$, and, assuming that the integral of the function $[a(t) + b(t)]$ is unity, N_{2R} (Raman term) is equal to 2. The result of Stolen *et al.* of $N_{2\infty}/N_{2R} = 0.82/0.18$ allows us to calibrate the ratio of fast to slow response ($\sigma = 6.07$).

For the PG FROG geometry[3,5] we use a Kerr-gate geometry in which a replica of the pulse gates itself. If the probe and the gate fields are of the same form, with the gate field delayed by τ and polarized at 45° with respect to the probe, the nonlinear polarization in the presence of the Raman effect can be obtained

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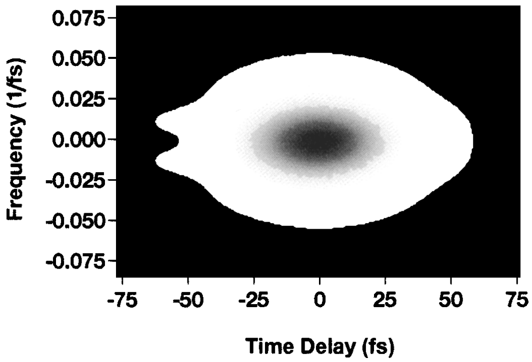

Fig. 1. PG FROG trace of a transform-limited Gaussian pulse with a FWHM of 25 fs. The material response function of fused silica, including the effects of the slow Raman terms, is used to generate the trace. The small features extending to negative delay times are the result of the Raman terms. If the material response were truly instantaneous, the trace would be a perfect ellipse. The trace background is set to black wherever the intensity is less than 10^{-4} of the peak in order to accentuate the slight distortion of the trace.

(again following Hellwarth[9]) as

$$\begin{aligned} P^{(3)}(t,\tau) = {} & \sigma E(t)|E(t-\tau)|^2 + E(t) \\ & \times \int_{-\infty}^{t} \mathrm{d}t' b(t-t')|E(t'-\tau)|^2 \\ & + E(t-\tau)\int_{-\infty}^{t} \mathrm{d}t'[2a(t-t') \\ & + b(t-t')]E(t')E^*(t'-\tau)\,, \end{aligned} \tag{2}$$

where the first term is the usual FROG signal field and the second and third terms are due to the slow response and correspond to the birefringence and the grating terms, respectively.

We use the exponentially damped sinusoid of Blow and Wood[10] as a reasonable approximation to the response function. Assuming identical functional forms of $a(t)$ and $b(t)$ (because the two functions are of similar magnitude and their sum is reasonably well approximated by this single function), we can write

$$a(t) = \frac{43}{57}\,\frac{{\tau_1}^2 + {\tau_2}^2}{\tau_1{\tau_2}^2}\,\exp(-t/\tau_2)\sin(t/\tau_1) \tag{3}$$

and $b(t) = 14a(t)/43$ (using the results in Table I of Ref. 11), where $\tau_1 = 12.2$ fs and $\tau_2 = 32$ fs.

The full FROG signal field, Eq. (2), when Fourier transformed and magnitude squared, becomes the FROG trace.[4] In Fig. 1 we see the PG FROG trace created by use of the signal field of Eq. (2) and the response of Eq. (3) for a Gaussian, transform-limited pulse with a full width at half-maximum (FWHM) of 25 fs. The small tails seen extending to negative delay times are a result of the slow Raman response of fused silica. Without the slow response this FROG trace is a perfect ellipse, without any such tails.[5]

The standard FROG pulse-retrieval algorithm, which explicitly assumes an instantaneous response, attempts to fit the Raman-induced features of the trace by modifying the retrieved pulse. As a result, it does not retrieve the correct pulse when a nonnegligible slow component of the response exists. Using Gaussian, transform-limited pulses with a FWHM of 10 elements on a 64-element array as input, we found that the standard algorithm retrieved pulses slightly longer than the actual pulses and slightly asymmetric. The amount of broadening is largest for pulses of 25-fs FWHM, as seen in Fig. 2. Longer pulses are not affected, because the slow response is short

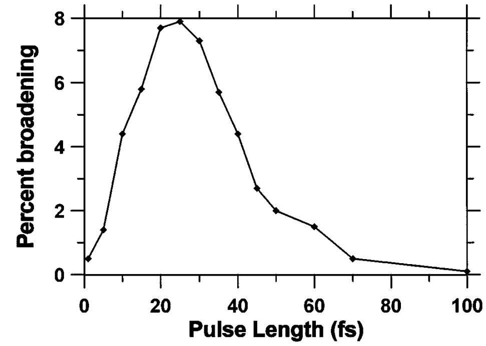

Fig. 2. Amount of temporal broadening in the pulse retrieved with the standard FROG algorithm that is due to the noninstantaneous Raman response of fused silica.

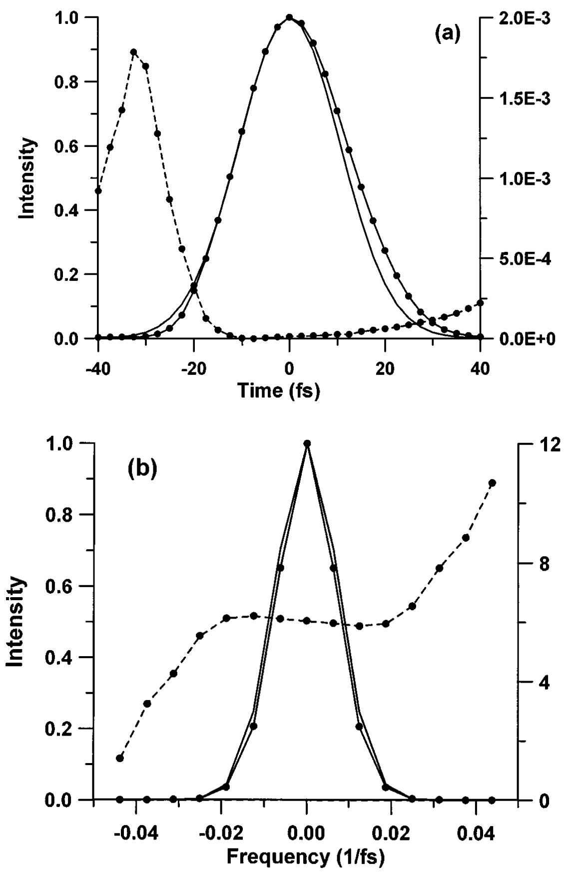

Fig. 3. Intensity and phase in (a) the time domain and (b) the frequency domain of the pulse retrieved (solid curve with dots) by the standard FROG algorithm from a FROG trace (Fig. 1) distorted by the Raman response of fused silica. The original pulse (solid curve) was a 25-fs FWHM transform-limited Gaussian pulse. The standard FROG algorithm retrieves a pulse that is 8% longer in its temporal FWHM and that has acquired some spectral cubic phase.

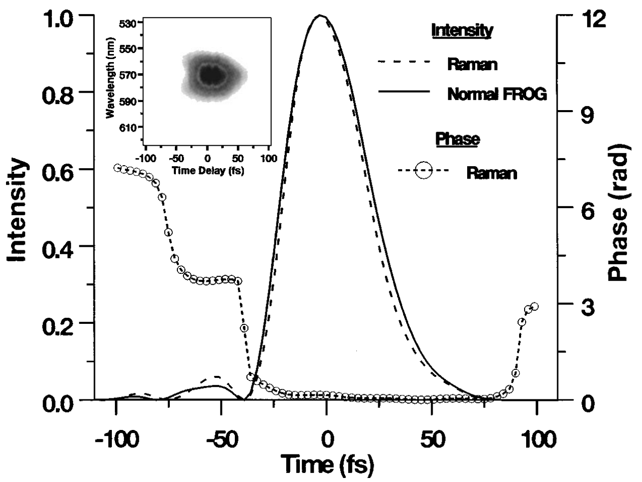

Fig. 4. Comparison of the pulse intensities derived by the normal instantaneous-response-based FROG algorithm and the algorithm modified to include the Raman response of fused silica. The Raman-aware algorithm achieved a lower error (0.00622 compared with 0.00733) and a shorter pulse (42.4 fs compared with 43.9 fs). The pulse phase is also shown. Inset: The PG FROG trace of the pulse. The tails seen in this trace are due mostly to residual third-order phase in the grating compressor, not to the Raman effect.

compared with the pulse length. Extremely short pulses are not affected so strongly because the ratio of energy to intensity decreases with pulse length, so that the contribution from the integrals in Eq. (2) decreases relative to that of the fast term.

The effects on a 25-fs pulse retrieved with the standard FROG algorithm are shown in Fig. 3. We see that the time-domain pulse intensity is slightly distorted, while the frequency-domain phase has acquired a cubic character. These results are typical of the effect of the Raman terms on the retrieved pulse.

To avoid limiting the accuracy of PG FROG for 10- to 60-fs pulses, we now include Raman effects completely in a modified pulse-retrieval algorithm. The use of generalized projections[6] allows us to use an arbitrary response function in the FROG algorithm. In this case the time-domain error function that is now minimized in the algorithm is

$$Z = \sum_{i,j=1}^{N} |E'_{\text{sig}}(t_i, \tau_j) - P^{(3)}(t_i, \tau_j)|^2. \tag{4}$$

Here $E'_{\text{sig}}(t, \tau)$ is the signal field after magnitude replacement by the experimental data and inverse Fourier transforming (just as in Refs. 4, 6, and 12), $P^{(3)}(t, \tau)$ is from Eq. (2), and the summation runs over all the N^2 points in the signal field array. This modified algorithm, in principle, exactly retrieves pulses even in the presence of Raman effects. We have tested this modified algorithm on several types of pulses, including pulses with complicated intensity and phase structure, and have found that in practice all these pulses are retrieved exactly. The price to be paid for this increased accuracy, however, is a decrease in speed. The modified algorithm runs much more slowly with the noninstantaneous terms: the number of calculations scales as N^3 rather than N^2 as in the purely instantaneous case.

We have also tested this modified algorithm on experimental data. The inset of Fig. 4 shows the PG FROG trace, made with fused silica as the nonlinear medium, of a pulse from an optical parametric generator pumped by an amplified Ti:sapphire laser. When using the standard instantaneous-response-based FROG algorithm to convert this trace, we obtain a pulse with a 43.9-fs FWHM and a residual rms error per pixel of 0.00733. With the modified algorithm, including the Raman response, the FWHM of the retrieved pulse is 42.4 fs, and the error drops to 0.00622, indicating better convergence when the modified algorithm is used. Experimental noise sets a lower limit on the obtainable error. The intensities derived by the two algorithms are compared in Fig. 4. The theory predicts only a 2.7% broadening for this pulse length rather than the 3.5% broadening observed here. This discrepancy is probably due to experimental noise. We observed a similar reduction of the retrieval pulse with for two other experimental traces of 42- and 34-fs FWHM.

The pulses discussed in this Letter have the same time scale as the Raman response; yet they could be retrieved because the response is known. The same concepts discussed here could, in principle, be used for the converse problem: to extract the response of a medium by using the knowledge of the pulse field. In other words, if fully characterized pulses are used in an experiment, an algorithm such as that described here may perhaps deconvolve out the ultrafast response of a medium, even though it is of the order of, or even shorter than, the pulses used to measure it.

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*Permanent address, Departamento de Física, Universidad Simón Bolívar, Apdo. 89000, Caracas, 1080A, Venezuela.

References

1. A. M. Levine, E. Ozizmir, R. Trebino, C. C. Hayden, A. M. Johnson, and K. L. Tokuda, J. Opt. Soc. Am. B **11,** 1609 (1994).
2. J. L. A. Chilla and O. E. Martinez, Opt. Lett. **16,** 39 (1991).
3. D. J. Kane and R. Trebino, Opt. Lett. **18,** 823 (1993).
4. R. Trebino and D. J. Kane, J. Opt. Soc. Am. A **10,** 1101 (1993).
5. K. W. DeLong, R. Trebino, and D. J. Kane, J. Opt. Soc. Am. B **11,** 1595 (1994).
6. K. W. DeLong, D. N. Fittinghoff, R. Trebino, B. Kohler, and K. Wilson, Opt. Lett. **19,** 2152 (1994).
7. R. H. Stolen, J. P. Gordon, W. J. Tomlinson, and H. A. Haus, J. Opt. Soc. Am. B **6,** 1159 (1989).
8. R. H. Stolen and W. J. Tomlinson, J. Opt. Soc. Am. B **9,** 565 (1992).
9. R. W. Hellwarth, Prog. Quantum Electron. **5,** 1 (1977).
10. K. J. Blow and D. Wood, IEEE J. Quantum Electron. **25,** 2665 (1989).
11. R. Hellwarth, J. Cherlow, and T.-T. Yang, Phys. Rev. B **11,** 964 (1975).
12. K. W. DeLong and R. Trebino, J. Opt. Soc. Am. A **11,** 2429 (1994).