# Femtosecond Kerr-lens autocorrelation

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An autocorrelation measurement of femtosecond laser pulse duration using the Kerr-lens mechanism is demonstrated. This technique can also be used as a sensitive and absolutely calibratable method for measuring ultrafast optical nonlinearities. A method that uses an electronic spectral-filtering scheme is proposed for determining the frequency chirp of pulses by interferometric autocorrelation. © 1997 Optical Society of America

The proliferation of ultrashort-pulse lasers has been accompanied by considerable progress in pulse diagnostics, including elaborate methods for determining the exact temporal behavior of the phase and the amplitude of the pulsed electric field.[1] In many practical cases, however, such detailed information is not necessary, and only the pulse duration ($t_p$) and the absolute magnitude of the frequency chirp need to be determined. For this purpose, second-order autocorrelation techniques have proved to be simple yet sufficient.[2] This type of autocorrelation requires a nonlinear-optical material with a response that exhibits a quadratic irradiance dependence:

$$I_{\mathrm{out}} = AI_{\mathrm{in}} + BI_{\mathrm{in}}^{\,2}\,, \tag{1}$$

where $I_{\mathrm{out}}$ and $I_{\mathrm{in}}$ are the output and the incident pulse irradiance, respectively. A process most commonly employed for this purpose is second-harmonic generation (SHG) within the low-pump-depletion limit. In that case, $I_{\mathrm{in}}$ represents the incident irradiance at the fundamental frequency ($\omega$) and $I_{\mathrm{out}}$ denotes the generated second-harmonic irradiance at $2\omega$. In addition, $A = 0$ (i.e., no linear background) and $B \propto |\chi^{(2)}|^2$, where $\chi^{(2)}$ is the second-order nonlinear-optical susceptibility. To obtain the autocorrelation, one splits the laser beam into two paths (e.g., with a Michelson interferometer) that have a variable relative delay ($\tau$). The exit irradiance of the interferometer is $I_{\mathrm{in}} = |E(t) + E(t+\tau)|^2$, where $E(t)$ is the instantaneous electric field of the laser pulse. The combined fields impinge upon the nonlinear medium to give an integrated output signal: $S(\tau) = \int I_{\mathrm{out}}(t)\mathrm{d}t$, where the integration limit is usually determined by the response time of the detection system.

The quadratic dependence in Eq. (1) can also be achieved with third-order nonlinearities $\chi^{(3)}$ involving nonlinear absorption or nonlinear refraction. In the low-irradiance limit, optical measurements involving either nonlinear absorption or nonlinear refraction give rise to a change of transmitted irradiance that varies quadratically with the incident irradiance. For example, two-photon absorption,[3] polarization Kerr gating,[4] and beam deflection induced by the optical Kerr effect ($n_2$) (Ref. 5) have been employed to yield pulse autocorrelations.

In this Letter a simple $\chi^{(3)}$ autocorrelator using the Kerr-lensing mechanism is demonstrated. In a Kerr-lens measurement such as Z scan,[6,7] the transmitted irradiance can be approximated as a quadratically varying function of incident irradiance, as in Eq. (1). One can assess the sensitivity of such a measurement by evaluating the change of transmittance $\Delta T[= I_{\mathrm{out}}/AI_{\mathrm{in}}$ in Eq. (1)] versus $I_{\mathrm{in}}$. Assuming an optical Kerr coefficient $n_2$ and a thickness $L$ for the nonlinear sample, we calculate $\Delta T$ as a function of the nonlinear phase shift, $\Delta\Phi_0 = (2\pi/\lambda)n_2 I_{\mathrm{in}} L$, using the models given by Refs. 6 and 7. These models assume a thin sample approximation, which requires that $L \le n_0 Z_0$, where $Z_0$ is the Rayleigh range of the focused Gaussian beam and $n_0$ is the linear refractive index of the material.[8] The results are plotted in Fig. 1 for an aperture (conventional Z scan) and an obscuring disk (eclipsing Z scan), both of which have transmittance of 0.02. The sample position is fixed at points corresponding to the peaks and valleys of the Z scans. These points are $Z = \pm 0.85 Z_0$ for the aperture[6] and $Z = \pm 0.5 Z_0$ for the disk,[7] where $Z$ is the distance from the focus. The dashed lines in Fig. 1 are linear fits with approximate slopes of $\pm 0.2$ for the aperture and $\pm 2$ for the disk. As expected, the calculation shows that a far-field disk has higher sensitivity.[7] More importantly, note that $\Delta T$ exhibits a large dynamic range while remaining linear in the time-averaged phase shift $\langle \Delta\Phi_0 \rangle$. This is important for obtaining an accurate and conveniently retrieved autocorrelation measurement. In Fig. 1 a comparison is made with the case of a two-photon absorber (2PA) placed at focus. Here, $\Delta T$ is plotted versus the time-averaged parameter $\langle Q_0 \rangle = \langle \beta I_{\mathrm{in}} L \rangle$ (solid line), where $\beta$ is the two-photon absorption coefficient.[6] A clear deviation from linearity (dashed line) is evident.

The Kerr-lens autocorrelation (KLAC) arrangement shown in Fig. 2 is nearly identical to that of a collinear SHG autocorrelator that uses a Michelson interferometer. We simply replace the SHG crystal with a Kerr medium (i.e., a semiconductor). The sample must be positioned at the peak or the valley of an appropriate Z scan, which requires that a partially obscuring aperture or (preferably) disk be placed before the far-field detector.

Figure 3 depicts rapid-scan autocorrelations of ~100-fs (FWHM) pulses from a Ti:sapphire cw self-mode-locked (75-MHz) laser obtained with the KLAC technique. The output of the interferometer was focused with an $f = 7.5$ cm lens to a spot size of $w_0 \approx 20\ \mu$m. The nonlinear medium in this experiment is 2-mm-thick polycrystalline ZnS placed at the

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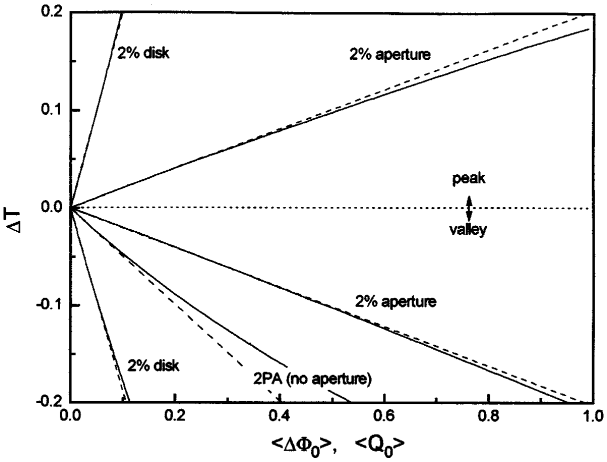


Fig. 1. Calculated transmittance change as a function of average nonlinear phase shift $\langle \Delta\Phi_0 \rangle$ assuming a 2% transmittance aperture with nonlinear sample position at $z = \pm 0.85 z_0$. Also shown is calculation for a 2% transmitting on-axis disk with the sample at $z = \pm 0.5 z_0$. The dashed curves are linear fits to the calculated curves. For comparison, the change of transmittance for a two-photon absorber placed at the focus is also plotted.

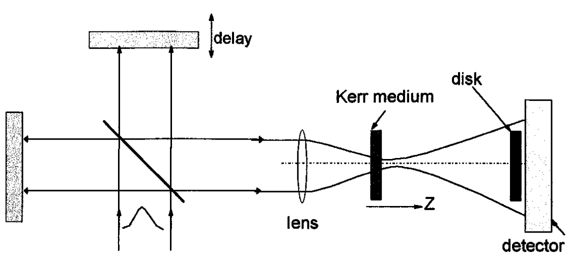


Fig. 2. Typical arrangement for a collinear Kerr-lens autocorrelator.

peak position. An obstruction disk yielding 2–3% transmission is placed in front of a silicon photodiode (Thorlabs, Inc., Model DET100). Figure 3(a) shows a KLAC trace obtained with an incident average power of 80 mW (per interferometer arm) superimposed upon a theoretical fit assuming a sech$^2$ temporal profile. Figure 3(b) shows two normalized KLAC traces measured at incident powers of 80 and 40 mW, revealing the linear dynamic range. It is worth noting that, in an eclipsing Z-scan geometry (i.e., when a disk is used),[7] linearity is maintained for transmittance changes of nearly 40%.

The choice of nonlinear material depends on the wavelength range and the laser pulse width. While it is desirable to have the largest $n_2$, the nonlinear response must have a turn-on time shorter than the pulse width. For femtosecond pulses the bound electronic Kerr effect in semiconductors is appropriate for this purpose. The optimum material is a semiconductor (or a dielectric) with a band gap just above the two-photon absorption wavelength ($E_g > h\nu/2$). This operating condition has been shown to give the largest value of $n_2$ without deleterious two-photon absorption.[9] For Ti:sapphire lasers ($\lambda = 700–900$ nm) ZnS is the most suitable semiconductor, whereas ZnSe and CdSe work best at $\lambda > 950$ nm and $\lambda > 1400$ nm, respectively. The optimum thickness of the nonlinear material depends on two factors: optimum focusing requires that $L \approx Z_0$, whereas avoiding pulse-broadening that is due to group-velocity dispersion necessitates that $L < Z_d = \pi c^2 t_p{}^2/[2\ln(2)\lambda^3|d^2n/d\lambda^2|]$. For ZnS at $\lambda = 800$ nm, $Z_d \approx 8$ mm for $t_p = 100$ fs. It is important to note that the thickness limitation set by group-velocity dispersion is less restrictive than length restrictions imposed by group-velocity mismatch in the SHG process.[10]

By Z-scan analysis,[6] KLAC can also be exploited to make sensitive $n_2$ measurements. Such a measurement is absolutely calibrated and highly accurate because it permits simultaneous determination of pulse width $t_p$, Rayleigh distance $Z_0$ (for locating the positions of the peak and the valley), and $\Delta\Phi_0$ (from $\Delta T$). An aperture is preferred for measuring $n_2$, whereas an eclipsing disk is more favorable for the autocorrelation measurements. Even though the disk has a larger slope $\Delta T/\Delta\Phi_0$, it is highly sensitive to the beam shape and to the amount of light that it transmits. With KLAC with an aperture, the nonlinear index of ZnS was measured as $n_2 \approx 6.5 \times 10^{-15}$ cm$^2$/W at $\lambda = 850$ nm.

The minimum power requirement for obtaining an autocorrelation is directly related to the smallest resolvable $\Delta T$. In the above experiment a $\Delta T$ of $\approx 0.1\%$ was resolved by standard averaging on a digital oscilloscope (Tektronix Model TDS-350). For an aperture this corresponds to an index change $\Delta n \approx 6 \times 10^{-7}$, which in turn gives a minimum irradiance of $10^8$ W/cm$^2$ for ZnS. Recalling the aforementioned laser parameters, we obtain a minimum average power $P_{\mathrm{ave}} \approx 6$ mW. Replacing the aperture with a disk improves the resolution to $P_{\mathrm{ave}} < 1$ mW. It must be noted that the beam quality of the laser affects only the sensitivity and not the shape of the autocorrelation trace. In other words, using KLAC as a pulse-width diagnostic tool does not necessarily require high beam quality if lower sensitivities can be tolerated.

Interferometric SHG autocorrelations have been used to obtain information about the laser pulse chirp.[11] For an irradiance temporal profile $f(t)$ the incident electric field is then given by $E(t) = \sqrt{f(t)}\cos[\omega t + \varphi(t)]$, where $\varphi(t)$ denotes the chirp. In

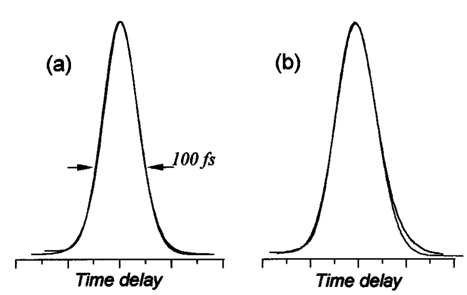


Fig. 3. Measured Kerr-lens autocorrelations of 100-fs Ti:sapphire laser pulses. (a) Comparison of measured data with calculated autocorrelation of a sech$^2$ pulse. (b) Two autocorrelation traces measured at average powers of 80 and 40 mW, indicating the dynamic linearity of the scheme.

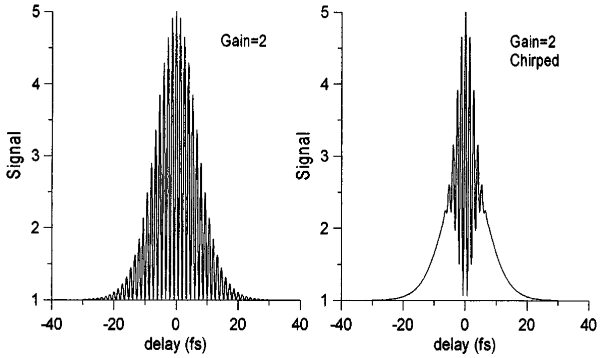


Fig. 4. Interferometric autocorrelation of 10-fs pulses calculated assuming that $G = 2$ for (a) unchirped and (b) chirped sech$^2$ pulses.

the SHG process the detected signal is then given by

$$S_{\mathrm{NL}}(\tau) = 1 + 2\int f(t)f(t+\tau)\mathrm{d}t + \int f(t)f(t+\tau) \times \cos(2\omega\tau + 2\Delta\phi)\mathrm{d}t + 2\int f^{1/2}(t) \times f^{3/2}(t+\tau)\cos(\omega\tau + \Delta\phi)\mathrm{d}t + 2\int f^{3/2}(t)f^{1/2}(t+\tau)\cos(\omega\tau+\Delta\phi)\mathrm{d}t\,, \tag{2}$$

where $\Delta\phi(t,\tau) = \phi(t+\tau) - \phi(t)$ and $\int f^2(t)\mathrm{d}t = 1$. In Eq. (2) the first integral represents the intensity autocorrelation and the remaining integrals are interferometric terms that contain information about the phase (i.e., chirp) of the laser pulse. Although accurate intensity autocorrelation can be easily obtained with the KLAC technique, interferometric measurements of the pulse chirp will be hindered by the presence of the linear background term indicated in Eq. (1). The difficulty arises from the fact that the transmitted optical field is now at the driving frequency ($\omega$) rather than the second harmonic ($2\omega$). The nonlinear signal cannot be distinguished from the linear term in Eq. (1) that contains the linear interferometric autocorrelation signal:

$$S_{\mathrm{lin}}(\tau) = 1 + \int [f(t)f(t+\tau)]^{1/2}\cos(\omega\tau + \Delta\phi)\mathrm{d}t\,. \tag{3}$$

The total detected signal is the weighted sum of the linear and the nonlinear contributions: $S(\tau) = S_{\mathrm{llin}}(\tau) + a\langle\Delta\Phi_0\rangle S_{\mathrm{NL}}(\tau)$, where $a$ is the appropriate linear slope depicted in Fig. 1. In a rapid-scan arrangement, delay is generated by vibrations on one of the interferometer mirrors so that autocorrelation is viewed in real time. We can understand this by noting that $\tau = t(2v/c)$, where $v$ is the maximum velocity of the mirror displacement and $c$ is the speed of light. Under this condition the interferometric fringes will appear in real time at a frequency $\Omega = \omega(2\nu/c)$, which can be made to fall in the radio frequency range. In such an arrangement spectral filtering can eliminate the linear interferometric term. The filtering process can be implemented either with electronic circuits (hardware) in real time or with Fourier analysis (software) after the autocorrelation is acquired. Similar to the scheme given in Ref. 12, by use of a band-pass–low-pass filter combination with a band-pass center frequency of $2\Omega$ the linear interferometric term at $\Omega$ can be rejected. Note that this filtering process eliminates the $\Omega t$ ($= \omega\tau$) term in $S_{\mathrm{NL}}$ as well. Here, by allowing for an adjustable gain ($G$) on the band-pass filter, we can write the total transmitted signal as

$$S(\tau) = 1 + 2\int f(t)f(t+\tau)\mathrm{d}t + G\int f(t)f(t+\tau)\cos(2\omega\tau + 2\Delta\phi)\mathrm{d}t\,. \tag{4}$$

Figure 4 shows $S(\tau)$ calculated for a $G = 2$ system, assuming (a) unchirped and (b) linearly chirped [$\varphi(t) = gt^2$; $g = 0.03\ \mathrm{fs}^{-2}$] pulses. The $G = 2$ case is particularly interesting because the autocorrelation of the unchirped pulse exhibits a fringe visibility (flat bottom) that makes it readily distinguishable from a chirped pulse. Such a filtering scheme can also be applied to SHG autocorrelations to facilitate chirp characterization.

In conclusion, a simple autocorrelation technique based on Kerr lensing has been demonstrated. The KLAC technique does not require phase or group-velocity matching and can be used as an inexpensive and simple alternative to SHG autocorrelation. This technique can also be used as a sensitive tool for measuring the optical Kerr effect by use of femtosecond lasers. A spectral filtering technique for obtaining interferometric KLAC traces was also proposed.

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