Spectral responsivity and efficiency of metal-based femtosecond autocorrelation technique

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Abstract

We have studied, for the first time experimentally, the efficiency and spectral responsivity of an autocorrelation technique that is based on second harmonic generation from metal surfaces. High-quality autocorrelation traces are obtained with pulse energies as small as only 64 pJ (intensity $\sim$6.5 MW/cm^2), resulting in an efficiency that is comparable with existing nonlinear-crystal-based autocorrelation techniques. Our scheme, however, has a number of significant advantages over most existing autocorrelation techniques because it is dispersion-free and provides automatic phase matching over an extremely wide spectral range. This technique has potential for temporally characterizing any light source with a super-broad spectrum, a task that cannot be accomplished with existing techniques.
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Recording of the auto- or cross-correlation functions provides the most convenient way for measuring the duration of sub-picosecond laser pulses [1,2]. Such measurements typically employ the Michelson or the Mach–Zehnder interferometric geometry and use a nonlinear crystal to generate second harmonic (SHG) from two temporally overlapped pulses. Correlation functions are obtained by recording the SHG intensity and allow one to deduce the duration of short pulses, assuming a certain pulse shape. Such a technique, although straightforward, has a number of limitations for characterizing femtosecond pulses. The most significant disadvantage stems from the phase-matching requirement, which limits the spectral bandwidth of pulse characterization. For example, an ultrashort pulse with a 40 nm bandwidth will in most cases suffer some cutoff at its

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spectral edge simply because a nonlinear crystal only optimizes SHG over a certain wavelength range determined by the orientation of the crystal c axis. Even if the crystal orientation is adjusted, phase matching restricts the wavelength range over which a nonlinear crystal can be used (at most over 1000 nm) [3]. Therefore, pulses at different central wavelengths (e.g., 800 vs 1500 nm) often require different types of crystals. In fact, no crystal is known to work efficiently for wavelengths shorter than 400 nm. Furthermore, dispersion in a nonlinear crystal itself introduces errors in short pulse measurements. Above does not account for the fact that nonlinear crystals are usually quite expensive, especially those made for special purposes (e.g., thin crystals needed for reducing dispersion).

In recent years, a technique based on two-photon absorption inside a photodetector has emerged for autocorrelation measurements [4–6]. This technique overcomes several limitations of the nonlinear-crystal-based technique as it does not require phase matching. However, it requires semiconductor material whose bandgap energy is larger than one-photon energy but smaller than two-photon energy ($h\gamma < E_g < 2h\gamma$). Therefore, different materials are still needed for different wavelengths. This feature may restrict the suitability of such a technique when pulses have a super-broad spectrum such as those produced from photonic-crystal fibers [7,8].

In this paper, we discuss a noncollinear autocorrelation scheme based on SHG from metal surfaces and experimentally demonstrate that it has significant advantages over previous techniques. The basic principle is as follows. Inside bulk metals, the inversion symmetry prevents SHG because the second-order susceptibility from electric dipoles vanishes [9,10]. However, this symmetry is broken at metal surfaces, and SHG can be generated from the surface atomic layers [9,10]. When a laser beam is incident onto a metal film from a certain angel, surface SHG will be generated collinearly along the direction of the reflected beam. However, when two beams are incident onto the metal sample with equal angle but from opposite direction, as shown in Fig. 1, surface SHG will be generated in the direction normal to the metal surface, bisecting the angle between the two inci-
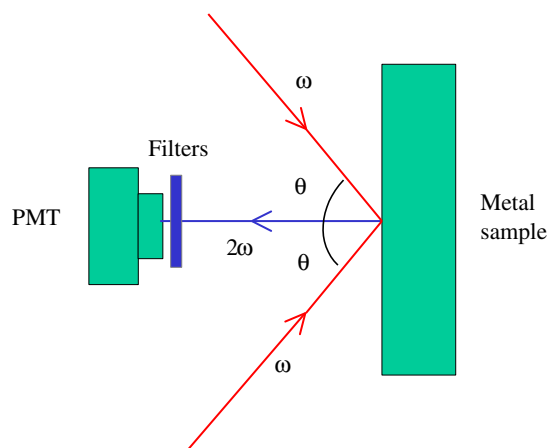

Fig. 1. Schematic illustration of a metal-based femtosecond autocorrelation technique.

dent beams. This SHG is dictated by the energy and momentum conservation laws, when one photon from each beam is absorbed simultaneously.

This surface SHG autocorrelation scheme provides a number of unique advantages. First, since SHG is generated from a few top surface layers, the phase-matching condition is automatically fulfilled. Second, the measurement is virtually dispersion free since only a few atomic layers are involved, and the reflected signal is collected (avoiding materials dispersion to transmit light). Third, SHG can be generated from a metal surface over an extremely broad wavelength range. In principle, the SHG signal can be produced from far infrared to deep ultraviolet, up to the plasma frequency of the metal used (e.g., about 10 eV for gold). This broad spectral bandwidth property is the most significant advantage over other existing techniques and it promises a unique way to characterize ultra-broad-band short pulses that may not be accomplished otherwise.

The potential disadvantage of the surface SHG technique is its possible low efficiency since the interaction length over surface layers is short. Although surface SHG has been used for pulse measurements in various materials, this issue of possible low efficiency has never been addressed before [11–13]. For example, incident fluence as high as 0.2 J/cm^2 was used to generate SHG from a semiconductor Si surfaces [11]. Furthermore,

although the most significant potential advantage for surface SHG autocorrelator is its broad spectral responsivity, this property has never been characterized experimentally. In this paper, we will address these two key issues (efficiency and broad spectral responsivity), for the first time experimentally, in the metal-based autocorrelation technique.

In this paper, we introduce a noncollinear autocorrelation setup based on SHG from metal surfaces that eliminates background offset. We demonstrate a high efficiency in generating autocorrelation SHG traces that is nearly comparable to most existing autocorrelation techniques. Spectral response of this metal-based autocorrelation technique is also measured and show significantly better spectral responsivity compared to nonlinear crystal based autocorrelation techniques.

Experimentally, we employ a femtosecond Ti:sapphire oscillator running at 800 nm central wavelength with a repetition rate of 94.3 MHz. The beam from the oscillator is split into two pulse trains using a 50/50 beam splitter such that each path carries nearly half the average power. The two beams are p polarized and focused onto the metal sample with two identical lenses. The second harmonic, generated through two-overlapping beams at the sample surface, is emitted in the normal direction, as shown in Fig. 1. A photomultiplier tube (PMT) is used to detect the surface SHG. Filters transmitting 400 nm but blocking 800-nm light are mounted right in front of the PMT to effectively cut off any fundamental leakage at 800 nm. In our experiment, the metal samples used are polycrystalline gold films. To obtain the autocorrelation trace, one of the beams is delayed relative to the other beam, and the photomultiplier records the intensity of the autocorrelation signal as a function of the delay between the two beams. A lock-in amplifier is used to enhance the signal-to-noise ratio. A typical autocorrelation trace obtained using the oscillator is shown in Fig. 2. As seen clearly, the autocorrelation traces obtained by our method exhibit a remarkably good signal-to-noise ratio. The pulse duration measured using our technique is further confirmed by regular autocorrelation measurements using a KDP nonlinear crystal. The efficiency of SHG depends on the inci-
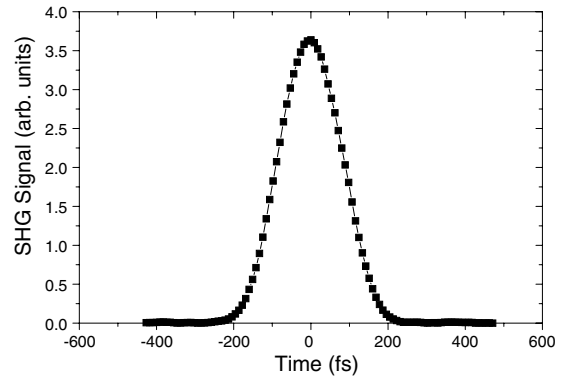

Fig. 2. A typical experimental autocorrelation trace obtained using surface SHG from gold.

dent angle of the fundamental beam (angle θ in Fig. 1). We find that the signal is readily detectable over a large angular range from 20° to 80°, with the peak efficiency between 65° and 70°. Typically, the time resolution of the autocorrelator at large incident angles can be improved by focusing the laser beam to a smaller spot on the metal surface. Our experience and calculations show that the scheme discussed here is suitable for measuring pulse duration from almost any femtosecond laser oscillator.

To ensure that the observed signal in the normal direction is indeed due to SHG from the gold surface, we measure the signal strength as a function of the average laser power from one beam. For two beams incident at 65°, this power dependence is shown in Fig. 3. The solid line is a quadratic fit to the data. The observed signal shows an excellent quadratic dependence on the incident beam power, as expected for any SHG signal, leaving no doubt about its origin. Furthermore, we can see from the figure that the autocorrelation SHG signal can be obtained with input average power level as low as 6 mW per beam from a 94.3 MHz repetition-rate oscillator. This value corresponds to a pulse energy of only 64 pJ and to an intensity level of about 6.5 MW/cm^2 in our experimental setup. Therefore, the SHG efficiency of this new technique is comparable to those based on using a bulk nonlinear crystal even though our technique enjoys significant advantages in terms of phase matching, dispersion, and broad spectral response,
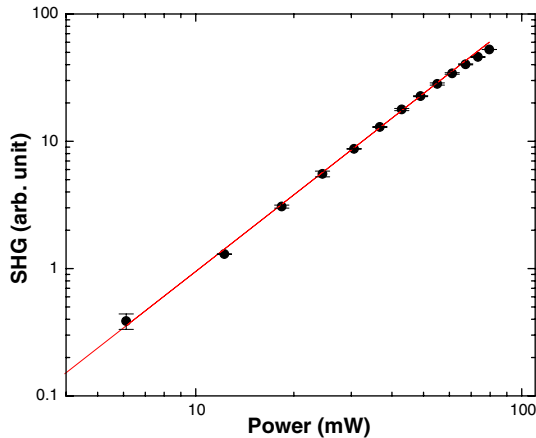
Fig. 3. Power dependence of the autocorrelation SHG signal generated in the normal direction of the gold sample. The solid line is a quadratic fit to the SHG signal. The data are slightly reduced at the highest intensities due to detector saturation.

Fig. 4. Spectral dependence of the autocorrelation SHG signal. The solid curve shows the calculated spectral response of a 100-μm thick type-I BBO crystal assuming that the crystal is oriented at the optimal phase-matching angle (29.18°) for 800 nm light.

as elaborated in the introduction. We expect that a clean SHG signal can be obtained even at lower pump power levels with additional efforts to enhance the signal-to-noise ratio.

As pointed out in the introduction, one of the key advantages of the metal-based autocorrelation technique is the much broader spectral range over which the SHG signal can be efficiently generated compared with other techniques. To demonstrate this feature experimentally, we try to run our Ti:sapphire oscillator over a range of central wavelengths. However, due to the spectral bandwidth limitation of the optics used in the oscillator, stable mode-locked pulses lase only over a 780–850 nm central wavelength range. Autocorrelation signals are recorded over this wavelength range, and Fig. 4 shows the wavelength dependence of the measured peak autocorrelation signals. To account for the variations in power, pulse duration, filter and detector responsivity, and focused spot size as the wavelength is changed, we normalized the autocorrelation SHG signal by a factor of $\tau \kappa \lambda_0^2 / P^2$, where τ denotes pulse duration, κ accounts for the detector and blue filter responsivity, λ_0 is the central wavelength of the pulse, and P the incident average power [6]. As shown in Fig. 4, the spectral response is relatively flat over the measured spectral range. For comparison, we also
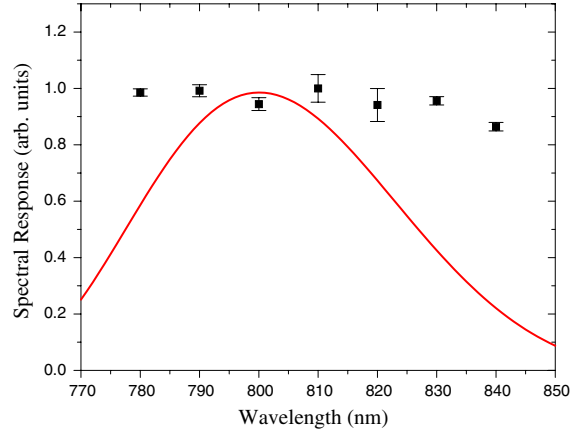
calculated the spectral response of a 100-μm thick type-I BBO crystal assuming that the crystal is oriented at the optimal phase-matching angle (29.18°) for 800 nm light according to [14]. The calculated curve is plotted against the experimental data. It is evident from the figure that our technique shows a significantly better spectral response compared to nonlinear-crystal-based measurements. Further tests over a broader spectral range can be carried out using different or broadly tunable light sources.

In summary, we have studied, for the first time experimentally, spectral responsivity and efficiency of an autocorrelation technique that is based on SHG from metal surfaces. High-quality autocorrelation traces are obtained with pulse energies as small as only 64 pJ (corresponding to an intensity of about 6.5 MW/cm^2), resulting in an efficiency comparable with current nonlinear-crystal-based measurements. However, the demonstrated scheme has significant advantages over the existing autocorrelation techniques inasmuch as it is dispersion-free, provides automatic phase matching, and is capable of responding over an extremely wide spectral range. This technique not only provides an alternative for current femtosecond pulse measurements, but also is likely to emerge as a strong candidate for temporally characterizing any light source with

a super-broad spectrum, such as those produced from photonic-crystal fibers [7,8].

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