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Autocorrelation Measurement of Femtosecond Optical Pulses Based on Two-Photon Photoemission in a Photomultiplier Tube

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The very sensitive intensity autocorrelation measurement of 15 fs, 800 nm pulses from a Ti:sapphire laser has been achieved using two-photon-induced photocurrent in a photomultiplier tube.

KEYWORDS: femtosecond, autocorrelation, photomultiplier tube, two-photon photoemission, ultrashort pulses

Recent progress in intensity autocorrelation measurements of ultrashort optical pulses based on two-photon-induced processes in photodetectors, such as photodiodes (PDs) and light-emitting diodes (LEDs), has made the pulse-width measurement of ultrashort pulses very simple and easy.[1–8] Virtually instantaneous two-photon interband transition in these semiconductor materials makes these methods very powerful particularly for the measurement of pulses shorter than 30 fs, in which the deterioration of the time resolution due to the group delay mismatch in a second-harmonic crystal becomes serious. The sensitivity of these methods is generally better than that of the conventional technique based on second-harmonic generation in a nonlinear crystal. However, more sensitive measurements are required in many areas of application of femtosecond optical pulses, such as pulse shaping and ultrafast spectroscopy. Kikuchi has reported sensitive autocorrelation measurements using a Si avalanche PD.[9]

Here, we present our recent results on the very sensitive autocorrelation measurement of optical pulses with a pulse width of 15 fs based on two-photon-induced photoemission in a photomultiplier tube (PMT). PMTs are very sensitive photodetectors because of the secondary-emission multiplication of photoelectrons. Two-photon-induced photoemission from photocathodes of PMTs has been observed,[10,11] and autocorrelation measurements of picosecond optical pulses have already been reported.[12,13] However, no studies have been reported on the femtosecond response of the two-photon-induced processes in PMTs, or on autocorrelation measurements of 800 nm light from Ti:sapphire lasers, which are widely used as femtosecond pulsed light sources.

In PMTs, electrons need to acquire energy which is greater than the sum of the band-gap energy and the electron affinity to be emitted into vacuum. Two-photon photoemission in PMT, therefore, can occur via a two-step process, where the first photon excites an electron from the valence band to the conduction band, and the second photon excites the electron above the vacuum level. Thus, measurements of ultrafast response time in PMTs are important for their use as nonlinear autocorrelation detectors. A finite response time of two-photon-induced processes in a type of PMT which is different to that used in the present study has already been observed.[14]

The experimental setup is as follows. The light source used in the experiments was a laboratory-built Kerr-lens mode-locked Ti:sapphire laser.[15] The laser cavity was dispersion-compensated by an intracavity silica prism pair. The cavity configuration was based on the design reported by Asaki *et al.*,[16] and pumped by an intracavity frequency-doubled Nd:YVO_4 laser with an output power of 5 W (Spectra Physics, Millennia V). The Ti:sapphire crystal had a path length of 4.5 mm. The pulse width, the wavelength, the average power, and the repetition rate of the output pulses were 15 fs, 800 nm, 600 mW, and 89 MHz, respectively. The output was attenuated by a reflective variable attenuator, and sent to a Michelson-type interferometer. Two beam splitters were used in the interferometer to balance the group-velocity dispersion in the two arms. The output of the interferometer was loosely focused by a concave mirror with a focal length of 200 mm onto the photocathode of a PMT. The beam size at the focus was 80 μm. The average power of the incident light was measured with a power meter at a delay time longer than 300 fs in order to eliminate effects of interference between the light of the two arms. The delay time of one arm in the interferometer was scanned by a translation stage.

We used a side-on type PMT with a reflection-mode photocathode (1P28, Hamamatsu Photonics). The cathode material of this PMT is Cs_3Sb (nominally Sb-Cs). This PMT has a one-photon response spectrum similar to that of a GaAsP PD, which was used by Ranka *et al.* in the autocorrelation measurement based on the two-photon-induced photocurrent in the PD.[4] Under low input intensity, this PMT is almost insensitive to 800 nm light, and has high sensitivity at 400 nm. A GaAsP PD was also used in the present experiment for comparison with the results obtained with the PMT. The PD was set in the same place as the PMT in the experimental setup.

Interferometric autocorrelation traces obtained with the GaAsP PD at two values of input power are shown in Fig. 1. Figure 1(a) shows an autocorrelation trace with a very good signal-to-noise ratio, which was obtained with 70 mW incident light. This autocorrelation trace was simulated very well with a 15 fs Gaussian input pulse. On the other hand, with incident light of 500 μW, autocorrelation traces obtained with the PD were not only very noisy but also deformed by the considerable contribution of the one-photon-induced photocurrent, as shown in Fig. 1(b).

With similar input power, a very good autocorrelation trace was obtained with the PMT. The result is shown in Fig. 2. The input power was 410 μW. Envelopes of a simulated autocorrelation trace of 15 fs Gaussian pulses are also shown in Fig. 2. No broadening of the pulse is observed in the figure, which shows that the two-photon processes in the PMT are sufficiently fast for autocorrelation measurements of 15 fs optical pulses. The ratio of the peak value to that of the baseline in the experimentally obtained trace is about 7:1, which is lower than the ratio of 8:1 for the ideal case. This reduction

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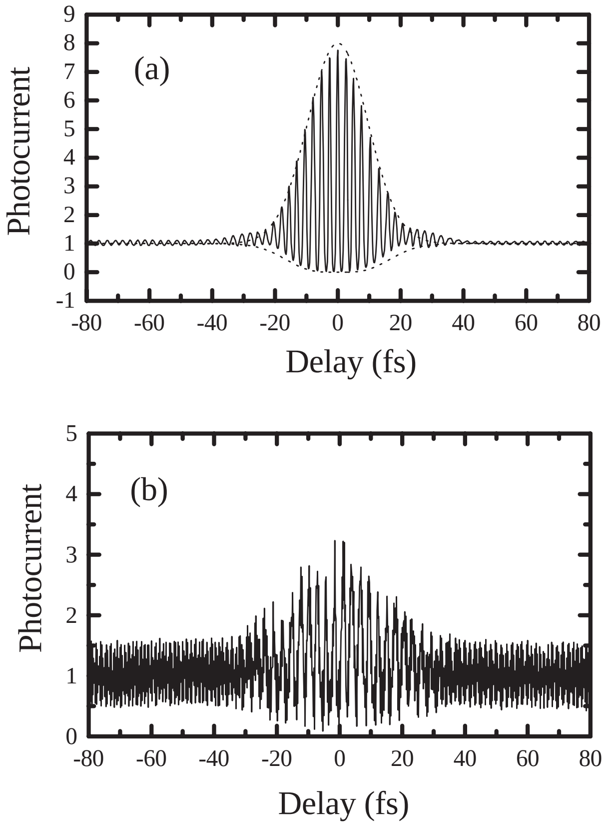

Fig. 1. Autocorrelation traces obtained with a GaAsP photodiode at different levels of incident light power: (a) 70 mW, and (b) 500 μW. Envelopes of an autocorrelation trace simulated with 15 fs Gaussian incident pulses are also shown in (a) by dotted lines.

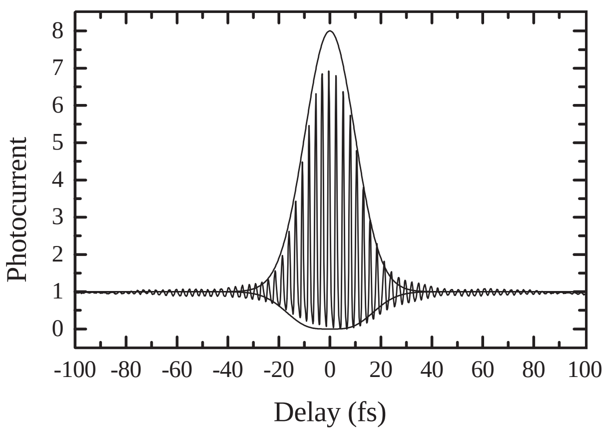

Fig. 2. Interferometric autocorrelation trace of 15 fs, 800 nm pulses from a Ti:sapphire laser obtained by the measurement of two-photon-induced photocurrent in a 1P28 PMT at input power as low as 410 μW. Simulated envelopes of the autocorrelation trace of 15 fs Gaussian pulses are also shown.

of the ratio is presumably due to the small contribution of the weak one-photon-induced photocurrent, which should have a peak-to-base ratio of 2:1. At lower input powers, smaller ratios of peak and base signal intensities were observed. At input powers higher than 500 mW, on the other hand, darkened spots were observed on the photocathode, presumably due to optical damage.

When the relative contribution of one-photon-induced processes to the signal at the baseline is denoted as x, and the

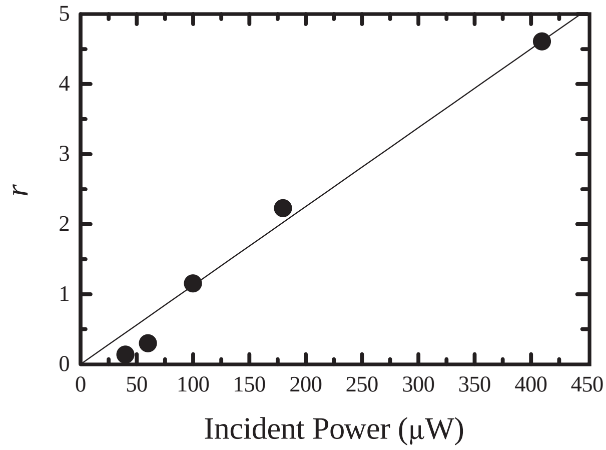

Fig. 3. Ratio of the contributions of two-photon-induced and one-photon-induced processes to autocorrelation traces obtained with the PMT.

two-photon contribution as $1 - x$, the peak-to-base ratio is calculated as $(8 - 6x) : 1$. Using this relation, the ratio, r, of the two-photon and the one-photon contributions;

$$r = \frac{1 - x}{x}, \tag{1}$$

in autocorrelation traces obtained with several incident powers are plotted as a function of incident power in Fig. 3. The plot exhibits a linear dependence of the ratio on the incident power, as expected. One-photon and two-photon processes are balanced at incident power of 90 μW, or single-beam power of 45 μW. The input power dependence of the photocurrent also showed a transition from linear dependence by a one-photon-induced process at lower input powers to quadratic dependence by a two-photon-induced process at higher input powers. Ranka *et al.*[4] reported the incident power dependence of photocurrent by a GaAsP PD for 100 fs incident pulses and cw light. The result obtained with 100 fs pulses showed quadratic dependence, and that obtained with cw incident light showed linear dependence. By extrapolating their results, the one-photon- and two-photon-induced signals are balanced around the incident power of 600 μW when the spot size on the detector is 80 μm. If we assume that the ratio of the one-photon- and two-photon-induced signal intensities is proportional to the incident pulse width, power of 600 μW corresponds to the double-beam power of 15 fs pulses at 180 μW. The result of the present study on the GaAsP PD shows that the one-photon- and two-photon-induced signals of the GaAsP PD are balanced around incident power of 250 μW. The small dispcrepancy between these values is probably due to the difference in the spectrum width of the 15 fs and the 100 fs pulses. Since the 800 nm light lies slightly below the band-gap energy of the PD material, 15 fs pulses have a larger spectral portion to which the PD is sensitive.

Now we compare the PMT and the PD as nonlinear detectors in autocorrelation measurements of weak light. Lower limits of the incident power in autocorrelation measurements are determined by two factors. The first is the signal-to-noise ratio of measured autocorrelation traces. PMTs are superior in this regard because of their high sensitivity, although lock-in detection or other signal-processing techniques can be used to relax the contrast. The other factor is the contribution of one-photon-induced processes. At very low incident power, two-photon-induced photocurrent is inevitably surpassed by one-photon-induced signal. The incident power at which the two contributions are balanced determines the inherent low power limit of the detector at the wavelength used. The present result shows that the PMT has an inherently low power limit which is about a factor of three lower than that of a GaAsP PD.

In summary, the very sensitive autocorrelation measurement of 15 fs, 800 nm optical pulses has been achieved using two-photon-induced photoemission processes in a PMT.

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