Autocorrelation measurement of femtosecond optical pulses using two-photon-induced photocurrent in a photomultiplier tube

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Abstract: Very sensitive intensity autocorrelation measurement of 15-fs, 800-nm pulses from a Ti:sapphire laser has been achieved by using two-photon-induced photocurrent in a photomultiplier tube. © 2000 Optical Society of America

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Recent progress in intensity autocorrelation measurements of femtosecond optical pulses based on twophoton-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[1-3]. Sensitivity of these methods are generally better than 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. Here, we present our recent results of very sensitive autocorrelation measurement of 15-fs optical pulses based on two-photon-induced photoemission in a photomultiplier tube (PMT).

PMTs are very sensitive photodetectors because of the secondary-emission multiplication of photoelectrons. Use of a PMT, therefore, as a nonlinear detector in autocorrelation measurements should lower the required input light power to a considerable extent. Autocorrelation measurements of picosecond optical pulses have already been reported[4, 5]. 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.

Experimental setup is described briefly in the following. The light source was a home-built Ti:sapphire laser generating 15-fs, 800-nm pulses at a repetition rate of 88 MHz. The output was attenuated by a reflective neutral-density filter, and sent to a Michelson-type interferometer. 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 μ m. The delay time of one arm in the interferometer was scanned by a translation stage. Two PMTs were used in the experiment. One was a side-on type PMT with a reflection-mode photocathode (1P28, Hamamatsu Photonics). The cathode material of this PMT is Sb-Cs (Cs₃Sb). The other was a head-on type PMT with a transmission-mode photocathode (R2557, Hamamatsu Photonics). The cathode material of it is bialkali (Na₂KSb). Both of these PMTs have one-photon response spectra 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[1]. Under low input intensity, these PMTs are almost insensitive to 800-nm light, and have high sensitivity at 400 nm. A GaAsP PD was also used in the present experiment for the comparison with the results obtained with the PMTs. The PD was put in the same place as the PMTs in the experimental setup.

Interferometric intensity autocorrelation trace with a good signal-to-noise ratio was obtained using a 1P28 PMT, as shown in fig.1. The input power, which was measured at a large delay time, was as low as 410 μ W. With this input power, traces obtained with the PD showed very large noise and were deformed by a considerable contribution of one-photon-induced photocurrent. Autocorrelation obtained by the PD with input power of tens of milliwatt level exhibited a clear trace, and was well simulated by Gaussian input pulses with a 15-fs width. Envelopes of a simulated autocorrelation trace of 15-fs Gaussian pulses are shown in fig.1. The experimentally obtained trace shown in the figure has a peak-to-base ratio of about 7:1, which is lower than the ratio of 8:1 for the ideal case. This reduction of the ratio is presumably due to a small contribution



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

of weak one-photon-induced photocurrent. Input power dependence of the photocurrent is shown in fig.2. The figure exhibits transition at around 270 μ W 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. This is consistent with the observed peak-to-base ratio at input power of 410 μ W.

Small asymmetry seen in the tails is probably due to a difficulty in the alignment with a PMT with a reflection-mode photocathode, which has thin wires in the path of the incident light. To eliminate this difficulty, the 1P28 PMT was replaced by an R2557 PMT, which has a transmission-mode photocathode, and the same measurement was performed. This type of PMT has a photocathode deposited on the inner surface of the entrance window, and better alignment can be achieved. Delay time dependence of photocurrent with this PMT is shown in fig.3. The spot size of the input beam was 80 μ m on the photocathode, and the input power was set at 350 μ W. Input power dependence of photocurrent, which is not shown here, was almost quadratic in the measured region of input power from 80 μ W to 600 μ W. In fig.3, tails are seen in the trace, which have a decay time of 270 fs. This suggests existence of an intermediate state in the two-photon



Fig. 2. Input power dependence of the photocurrent of a 1P28 photomultiplier tube. It shows transition from linear to quadratic dependence.



Fig. 3. Time-delayed two-photon-induced photocurrent trace by an R2557 photomultiplier tube. The tails show 270-fs relaxation of the intermediate state of the two-photon transition.

transition, and the decay time observed here is attibuted to the relaxation time of the intermediate state.

Photoelectrons are emitted from the photocathode of a PMT when the incident photon energy exceeds the sum of the bandgap energy and the electron affinity of the photocathode material, and the excited electron gets energy above the vacuum level. It has been reported that the bandgap energies of Cs_3Sb and Na_2KSb are 1.6 eV and 1.0 eV, respectively, although the sum of the bandgap energies and the electron affinities are 2.0 eV for both materials[6]. These facts explain the observed results. Since the 800-nm incident light has photon energy of 1.55 eV, it can induce interband transition with one photon in Na_2KSb , and electrons reach the vacuum level by two-photon absorption via a two-step process. Thus, PMTs without one-photon photocurrent response do not guarantee lack of one-photon absorption. On the other hand, in Cs_3Sb , no one-photon-induced transition is possible, and photoemission is induced by a direct two-photon process. In photodiodes and LEDs, however, photocurrent is expected to be generated whenever interband transition takes place. The small peak-to-base ratio (5.5:1) of the trace shown in fig.3 is attributed to existence of another relaxation process with a longer decay time in the intermediate state, which should raise the base level.

In summary, very sensitive autocorrelation measurement of 15-fs, 800-nm optical pulses has been achieved by using two-photon-induced photoemission processes in a PMT. Ultrafast relaxation of electrons excited in an intermediate state of the two-step two-photon transition was observed in a different type of PMT with the same measurement setup.

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