Autocorrelator for the measurement of a quasi-continuous-wave laser beam sinusoidally modulated at 17 THz

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Received 13 August 2006; received in revised form 1 September 2006; accepted 8 September 2006
Available online 7 November 2006

Abstract

The development of an autocorrelator, using a photomultiplier as a nonlinear optical detector, for the measurement of a quasi-continuous-wave laser beam in the milliwatt range, is described. The contribution of the two-photon absorption relative to the one-photon absorption was investigated for the Sb–Cs photocathode used in the photomultiplier by means of the Z-scan method. This autocorrelator was applied to the measurement of the intensity of a laser beam sinusoidally modulated in the terahertz range.

1. Introduction

A new “terahertz” technology has been developed for material science, medical/biological imaging, and ultrafast communication. In order to generate a terahertz wave, it is necessary to develop a laser source that emits at two different frequencies for the generation of a beat signal comprised of a sinusoidal wave in the terahertz range. For example, a nonlinear optical crystal can be installed in a high-finesse cavity, to generate a beat signal by coherent interaction with a pump source [1]. A continuous-wave (CW) Raman laser arising from molecular hydrogen in a high-finesse cavity for generating a Stokes beam, the frequency of which is determined by a Raman shift frequency (587 cm\textsuperscript{-1} for the rotational transition and 4155 cm\textsuperscript{-1} for the vibrational transition) has recently been reported. Thus, it should generate a sinusoidal wave in the terahertz range [2]. In order to measure the beat signal, it is necessary to use an autocorrelator with a femtosecond resolution. However, the peak power of the laser is in the milliwatt range, and, therefore, a sensitive as well as an ultrafast autocorrelator is essential for the measurement of the modulated beam. An autocorrelator based on the two-photon absorption of a photomultiplier photocathode has been developed for the measurement of a femtosecond pulse [3]. This approach does not require phase matching and is easily used in routine work [4–7].

In this study, we investigate the two-photon response of the Sb–Cs photocathode in a photomultiplier using the Z-scan method, in which the two-photon signal of the CW laser beam was measured by changing the distance from the beam waist, in order to change the intensity of the light without changing the flux of the beam. In fact, we were able to construct a sensitive autocorrelator that is capable of measuring a sinusoidal wave modulated at 17 THz by the stimulated Raman scattering of molecular hydrogen in a high-finesse cavity.

2. Experimental section

The nonlinear response of the photocathode was evaluated using the experimental apparatus shown in Fig. 1. A Ti:sapphire laser (Coherent, MBR-110, 450 mW) was used as a tunable CW laser in the wavelength range from 770 to 830 nm. The intensity of the laser beam was adjusted by means of a polarizer and a neutral density

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1468-6996/S - see front matter © 2006 Published by NIMS and Elsevier Ltd.
doi:10.1016/j.stam.2006.09.005
(ND) filter. A photomultiplier comprised of an Sb–Cs photocathode (Hamamatsu, 1P28, 190–650 nm) was used in this study. This photomultiplier has sufficient sensitivity for a single photon below 650 nm and has no sensitivity in the near-infrared region above 700 nm [8]. The Ti:sapphire laser beam was focused onto a photocathode using a lens with a focal length of 5 cm. The photomultiplier was moved by means of a translational stage (Sigma-koki, CLX-N610), the displacement of which can be directly monitored on the display of a computer. The photomultiplier was replaced with a beam profiler (OPHIR, Beamstar-V), and the beam profile at the photocathode was recorded. The signal from the photomultiplier was measured with a digital oscilloscope (Tektroniks, TDS380P, 400 MHz).

3. Results and discussion

Fig. 2 shows Z-scan plots of the signal from the photomultiplier. The laser wavelength was adjusted at 770, 792, and 830 nm. The radius of the beam was ca. 3 mm at the lens and was focused to ca. 20 μm at the beam waist. The baseline is slightly shifted from the zero level by the one-photon response of the photocathode. The signal arising from the two-photon response increases with decreasing beam radius in the vicinity of the beam waist, providing a peak with a Lorentzian shape, when the Z-scan method is used.

In the Z-scan method, the signal intensity arising from one- and two-photon absorptions, $S(\zeta)$, can be described as follows [9].

$$S(\zeta) = \sigma^{(1)}P + \sigma^{(2)}P^2 \left(1 + \frac{\zeta}{b^2}\right), \quad (1)$$

where $\sigma^{(1)}$ and $\sigma^{(2)}$ are the cross sections of one- and two-photon absorptions, respectively, $P$ is the peak power of the laser, and $\zeta$ is the parameter, defined as $\zeta = 2z/b$, in which $z$ is the distance from the beam waist to the photocathode surface, and $b$ is the confocal parameter. The ratio of the signals arising from one-photon (first term) and two-photon (second term) processes can then be calculated using Eq. (1). The signal arising from the one-photon process is independent of the parameter, $\zeta$ and can be measured as a baseline shift, as observed in Fig. 2. On the other hand, the signal arising from the two-photon process can be measured by fitting the observed data to a Lorentzian curve calculated using the second term of Eq. (1). It is suggested that the two-photon signal is three times larger than the one-photon signal for accurate measurement of the temporal profile. In this case, the ratio of the positive signal (positive peak signal measured from the baseline) and the negative signal (negative peak signal measured from the baseline) in the autocorrelation trace, $R$, becomes 6, which can be calculated by taking into account the contribution (3:1) of the two-photon response ($R = 8$) and the one-photon response ($R = 1$). From the observed data shown in Fig. 2, the minimum intensity of the laser, at which the ratio of the two-photon to one-photon signals is $3$, is $4.5 \times 10^3$, $4.1 \times 10^3$, and $3.4 \times 10^3$ W/cm$^2$ at 770, 792, and 830 nm, respectively. These results suggest that the contribution of the two-photon process increases with increasing laser wavelength, thus improving the sensitivity of the autocorrelator. The data also suggest that the intensity modulation arising from the beat signal of the fundamental (792 nm) and Stokes (830 nm) beams can be measured at an output power of 10 mW using an autocorrelator consisting of the Sb–Cs photocathode in the photomultiplier.

Fig. 3 shows a block diagram of the fringe-resolved autocorrelator developed in this study. The laser beam was split into two parts, which were recombined with each other after the beams were reflected by two pairs of retro reflectors in a Michelson interferometer. A piezo actuator (Piezosystem Jena, PX-100) was used, to translate one pair of the retro reflectors by 100 μm. The beam was aligned in collinear and focused onto the photocathode of a photomultiplier (1P28) using a lens with a focal length of 10 cm. The signal was measured by means of a digital oscilloscope (TDS380P).
Fig. 4 shows a fringe-resolved autocorrelation trace measured for a laser beam that is sinusoidally modulated at 17 THz. This result was obtained as the result of the beat arising from the fundamental beam and the Stokes beam induced by the stimulated Raman scattering of hydrogen in a high-finesse cavity [3]. The peak power of the modulated beam was ca. 10 mW (2.6 × 10^7 W/cm^2 at the beam waist). The ratio of the intensities for the positive and negative signals in the autocorrelation trace is ca. 4, which is slightly less than the value of 6, predicted by theory at an intensity of 4.1 × 10^3 W/cm^2. The small value might arise from a smaller intensity at the beam waist on the photomultiplier photocathode (2.6 × 10^7 W/cm^2) or an insufficient overlap of the two beams. It should be noted that the fringe spacing between the peaks is calculated to be 2.7 fs from the wavelengths of the fundamental and Stokes beams. Therefore, the separation of the sinusoidal signal is 57 fs, providing a modulation frequency of 17 THz. This result indicates that our system has enough time resolution for detection of THz light modulation.

4. Conclusion

In conclusion, a minimum laser intensity required for recording a two photon absorption of a photomultiplier photocathode (1P28) was determined to be 4.5 × 10^3 W/cm^2 using the Z-scan method. In fact, we constructed a sensitive autocorrelator for a quasi-CW laser and confirmed that the apparatus can be used for the measurement of a sinusoidally modulated (17 THz) beam formed as a signal beat of the fundamental beam and the Stokes beam generated by stimulated Raman scattering.

Acknowledgments

The authors thank Tomohiro Uchimura for technical assistance in this study. This work was supported by Grants-in-Aids for Scientific Research and for the 21st Century COE Program, “Functional Innovation of Molecular Informatics”, from the Ministry of Education, Culture, Science, Sports and Technology of Japan.

References

[1] K. Imai, K. Kawase, J. Shikita, H. Minamida, H. Ito, Appl. Phys. Lett. 78 (2001) 1026.
[2] M.R. Stone, M. Naftaly, R.E. Miles, I.C. Mayorga, A. Malcoci, M. Mikulics, J. Appl. Phys. 97 (2005) 103108.
[3] K. Ihara, C. Eshima, S. Zaitsu, S. Kamitomo, K. Shinzen, Y. Hirakawa, T. Imasaka, Appl. Phys. Lett. 88 (2006) 074101.
[4] W.R. Bennett Jr., D.B. Carlin, G.J. Collins, IEEE J. Quantum Electron. 10 (1974) 97.
[5] Y. Takagi, T. Kobayashi, K. Yoshihara, S. Imamura, Opt. Lett. 17 (1992) 658.
[6] T. Hattori, Y. Kawashima, M. Daikoku, H. Inouye, H. Nakatsuka, Jpn. J. Appl. Phys. 39 (2000) L809.
[7] J.M. Roth, T.E. Murphy, C. Xu, Opt. Lett. 27 (2002) 2076.
[8] T. Hattori, Y. Kawashima, M. Daikoku, H. Inouye, H. Nakatsuka, Jpn. J. Appl. Phys. 39 (2000) 4791.
[9] K. Ihara, S. Zaitsu, T. Imasaka, Rev. Sci. Instrum. 76 (2005) 026109.