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Letter

Constructive and Destructive Two-Pulse Excitation Investigated with a White-Light Michelson Interferometer

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Linear propagation of two pulses through methanol solution of aluminum phthalocyanine chloride is investigated using a modified white-light Michelson interferometer. The observed coherence time of the white light is 6 fs, and the separation between the two-excitation pulses is set to about 10 fs. The excitation is dependent on the phase-relation between the two pulses. We have observed an enhancement of the excitation when the two pulses are in-phase and strong suppression of the excitation when the two pulses are out of phase by π.

Key words: two-pulse excitation, Michelson interferometer, phase control, ultrashort pulse propagation, Bloch vector

1. Introduction

In recent years the manipulation of quantum systems with phase-controlled shaped pulses has attracted much attention.[1-4] In order to observe both the amplitude and the phase of light wave, we have to use interferometric method. Fourier transform spectrometers (FTS), which are common in infrared regions, use Michelson interferometers. In almost all the commercially available FTS's the sample is inserted on the output side of the Michelson interferometer, and the autocorrelation interferogram of the transmitted light is obtained. The absorption spectrum is then given by the Fourier transform of the autocorrelation interferogram. To obtain the phase information of the transmitted light, we have to insert the sample into only one arm of the Michelson interferometer and obtain not only the autocorrelation interferogram of the input light but also the cross-correlation interferogram between the input light and the transmitted light through the sample.

White-light Michelson interferometers have been proved to be useful in observing the deformation of light pulses by the passage through laser cavities, optical components and various kinds of materials.[5-9] We recently reported on an observation of linear propagation of light using a white-light Michelson interferometer in the visible and near infrared region.[10-12] In that experiment we obtained two kinds of interferograms: one was the autocorrelation of the input light, and the other was the cross-correlation between the input light and the light transmitted through a sample where the sample was inserted in only one arm of the white-light Michelson interferometer. The autocorrelation interferogram $C_A(\tau)$ is written as

$$C_A(\tau)=\langle E^*(t)E(t+\tau)\rangle , \tag{1}$$

where $E(t)$ is the electric field of the white-light, and $\langle\ \rangle$ denotes time average. In the linear response approximation, the electric field of the transmitted light, $E'(t)$, through the sample becomes

$$E'(t)=\int_{-\infty}^{\infty} dt' h(t-t')E(t') , \tag{2}$$

where $h(t)$ is the response function of the sample. The cross-correlation interferogram, $C_C(\tau)$, between the input light, $E(t)$, and the output light, $E'(t)$, from the sample is written as

$$C_C(\tau)=\langle E^*(t)E'(t+\tau)\rangle . \tag{3}$$

Substituting Eq. (2) into Eq. (3), we obtain $C_C(\tau)$ as

$$C_C(\tau)=\int_{-\infty}^{\infty} d\tau' h(\tau-\tau')C_A(\tau) . \tag{4}$$

By comparing Eqs. (2) and (4), we can say that if the waveform of the input light to the sample is the autocorrelation, $C_A(\tau)$, then the waveform of the output from the sample is the cross-correlation, $C_C(\tau)$. Therefore, in the linear regime we can see the deformation of ultrashort light pulses by the propagation through samples using a white-light Michelson interferometer. Here, we can consider one arm of the Michelson interferometer as the signal arm where the 'signal light' passes through and the other arm as the reference arm where the 'reference beam' passes through.

The white light or broad-spectral light has a very short coherence time which is inversely proportional to the spectral width. Therefore the white light can be considered as a random series of ultra-short light pulses with widths of the coherence time. The white light Michelson interferometer effectively picks up only one pulse by correlating the light fields from the two arms. This is the reason we can see the ultra-short pulse response using the white-light Michelson interferometer.

In the present experiment we split the signal arm into two arms thus obtaining a modified Michelson interferometer, so that we can observe two-pulse excitation. A schematic diagram of the experiment is shown in Fig. 1. The input light source is a normal incandescent lamp with power consumption of 650 W. The unpolarized light from the lamp is guided to a vibration isolated bench through a

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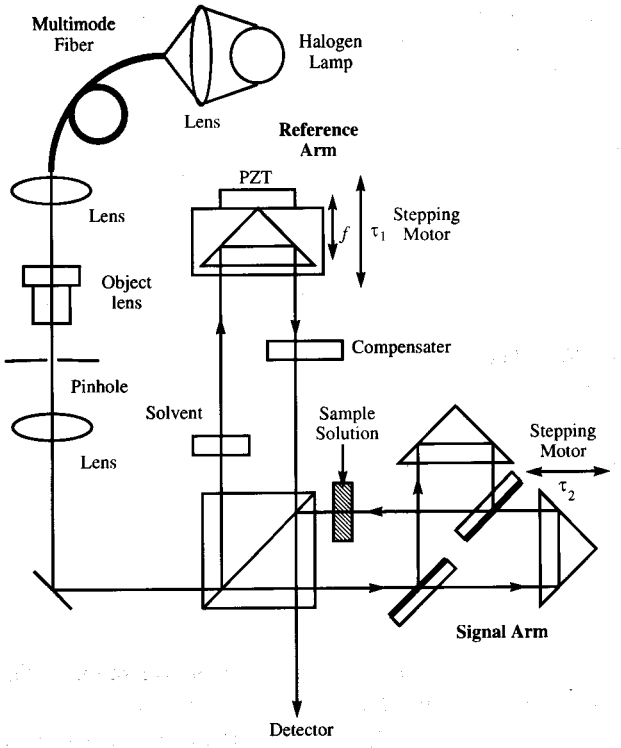

Fig. 1. Schematic diagram of the modified white-light Michelson interferometer for the observation of two-pulse excitation.

multimode optical fiber with a core diameter of 0.9 mm. We use the large core-diameter fiber to utilize as much light power as possible and to increase the signal-to-noise ratio in the measurement of the interferogram of strongly light-absorbing samples. The light output from the fiber is collimated by a lens and is passed through the modified Michelson interferometer. At the output of the interferometer, the two beams from the signal arms and the beam from the reference arm are made to spatially overlap with very high accuracy. In order to compensate for the dispersion caused by passing through extra glass material in the signal arm, we insert a compensator (a glass plate) in the reference arm. The path length of the reference arm is modulated at $f \simeq 1$ kHz by a piezoelectric actuator with an amplitude of 250 nm, and the output light intensity from the interferometer is detected by a photomultiplier tube and is measured with a lock-in amplifier. Interferograms are obtained by scanning the path length, or the delay time, of the reference arm, while the delay time between the two arms of the signal arms is set at a fixed value. Here the delay time between the two signal arms corresponds to the time-separation between the two excitation pulses.

The sample is aluminum phthalocyanine chloride (AlCl-Pc) in menthanol. The absorption spectrum in the visible region is composed of a large peak and two small peaks as shown in Fig. 2 where the AlCl-Pc concentration is 1.3×10^{-5} M. We first observe one-pulse propagation, where one of the two signal arms is blocked. In the measurement of the autocorrelation the same type of glass cells (inner width 2 mm) containing only the solvent are inserted into

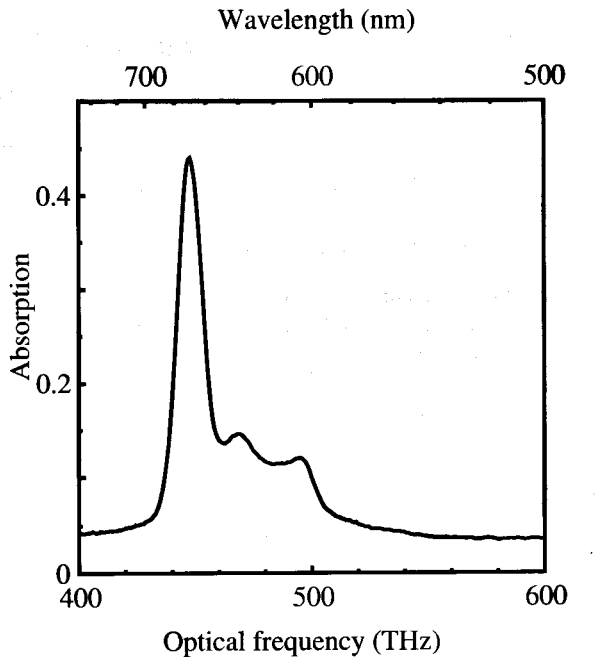

Fig. 2. Absorption spectrum of aluminum phthalocyanine chloride in methanol. The concentration is 1.3×10^{-5} M.

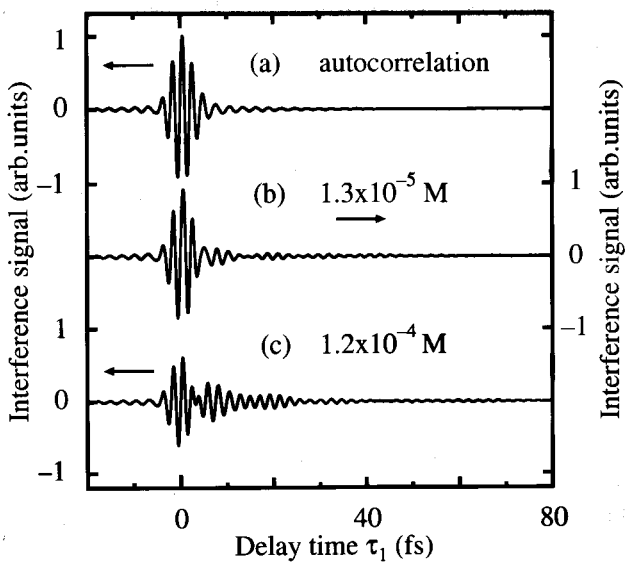

Fig. 3. Interferograms obtained with a Michelson interferometer: (a) autocorrelation of the incident white-light, (b) and (c) cross-correlation for single-pulse excitation, the concentration of AlCl-Pc is 1.3×10^{-5} M and 1.2×10^{-4} M, respectively.

both the signal and reference arms. The solvent is transparent in the visible and near infrared region. We obtain cross-correlation by replacing the solvent in the signal arm with the sample solution. In this way, the change in the optical path length due to the glass cell and the solvent is compensated for. Figure 3(a) shows the autocorrelation or 'input pulse,' and Figs. 3(b) and (c) show the cross-correlation or 'transmitted pulse' through the sample solution where the concentration of AlCl-Pc is 1.3×10^{-5} M and $1.2\times$

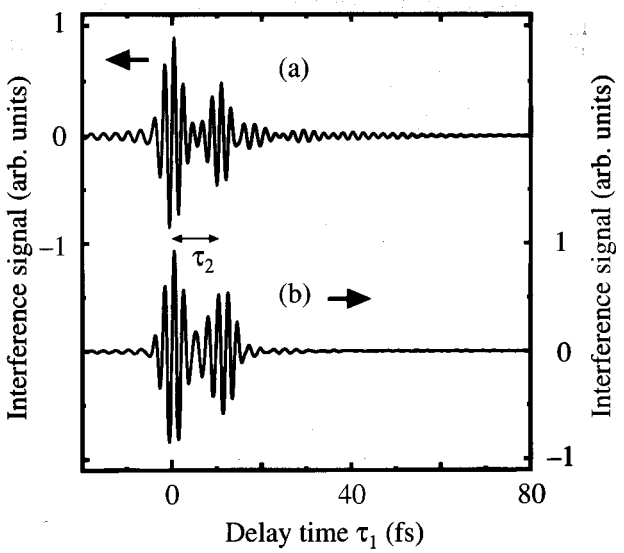

Fig. 4. Interferograms obtained with the modified Michelson interferometer: (a) cross-correlation for in-phase two-pulse excitation, (b) cross-correlation for out of phase two-pulse excitation, the concentration of AlCl-Pc is 1.3×10^{-5} M.

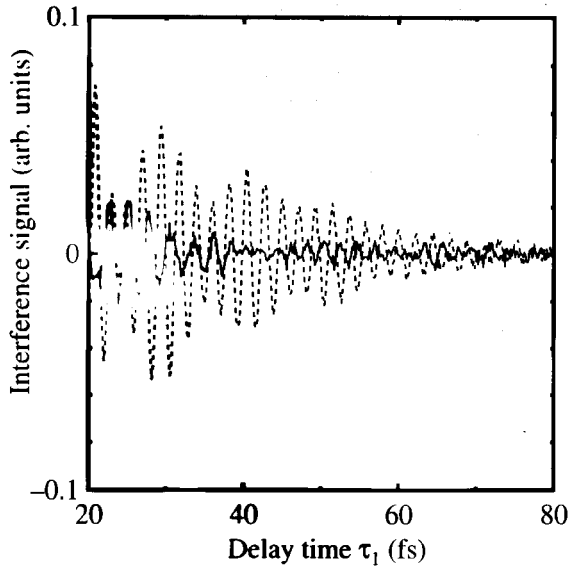

Fig. 5. Comparison of the free-induction decay signal between in-phase (dotted curve) and out of phase (solid curve) two-pulse excitation.

10^{-4} M, respectively. The width of the autocorrelation interferogram, 6 fs, is limited by the spectral response of the photodetector. In the cross-correlation the slowly decaying oscillation on the trailing side of the peak is the optical free-induction decay (FID) signal which is emitted from the polarization induced by the 'input pulse.' The beating of the FID signal is caused by the interference among components which correspond to the absorption peaks shown in Fig. 2. The main beat frequency ~20 fs agrees with the farthest absorption-peak separation ~50 THz.

The measurement of the two-pulse excitation is performed by setting the path-length difference or 'delay time' between the two signal arms at a fixed value and scanning the delay time of the reference arm. The path-length difference between the two signal arms corresponds to the time separation between the two excitation pulses. Since we wish to observe the phase dependence between the two excitation pulses, we keep the fluctuation of the path-length difference within a fraction of a wavelength of light during one scan of the delay time of the reference beam by carefully isolating the vibration of the optical bench and avoiding the air flow around the interferometer.

Figure 4(a) shows the cross-correlation for the two-pulse excitation where the pulse separation between the two excitation pulses is set at 10.5 fs and the concentration of AlCl-Pc is 1.3×10^{-5} M. In this case the first and the second pulses are in-phase, so that the polarization is produced constructively by the two excitation pulses to induce a large FID signal after the second pulse. On the other hand, Fig. 4(b) shows the cross-correlation for the same two-pulse excitation with almost the same two-pulse separation but the two excitation pulses are out of phase by π. Here, the polarization induced by the second pulse is out of phase by π with respect to that induced by the first pulse, and the two polarizations interfere destructively to produce no polarization and no FID signal after the second pulse. We note that the FID signal by two-pulse excitation is larger when the two pulses are in-phase, but it is smaller when the two pulses are out of phase by π, in comparison to that of one-pulse excitation for the same AlCl-Pc concentration shown in Fig. 3(b).

In the linear regime, any material response can be explained both in time domain and in frequency domain. But the response to the above two-pulse excitation is more simply elucidated in the time domain by using the optical Bloch vector for two-level systems. In a rotating frame, the Bloch vector is rotated by θ by the first excitation pulse. If the second pulse is in phase, then the Bloch vector is rotated by θ by the second pulse around the same axis so that it is rotated by a total of 2θ from the beginning. But if the second pulse is out of phase by π with respect to the first pulse, the Bloch vector is rotated by $-\theta$ by the second pulse so that it is rotated by $\theta-\theta=0$ in total from the beginning, and no excitation is left after the second pulse.

In Fig. 5 the FID signal parts of the interferograms are compared between the in-phase constructive two-pulse excitation and the out of phase destructive two-pulse excitation. The polarization induced by the first pulse decays by a factor $\eta=\exp(-\tau_{12}/T_2)$ until the arrival time of the second pulse, where T_2 is the dephasing time and τ_{12} is the separation between the two excitation pulses. Therefore, the most effective destruction of the polarization occurs if the amplitude of the second pulse is attenuated by η compared with the first pulse, as is shown in Fig. 4(b).

In summary, we have demonstrated that constructive and destructive two-pulse excitation can be investigated

with a modified white-light Michelson interferometer. If we replace the white light with phase-tailored light pulses, the quantum control by the phase-tailored pulses can be monitored by observing the FID signal with the Michelson interferometer.

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