Frequency-domain interferometer for femtosecond time-resolved phase spectroscopy

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A new femtosecond time-resolved interferometer was developed that utilizes interference fringes in the frequency domain to obtain simultaneously difference phase spectra (DPS) and difference transmission spectra with a multichannel spectrometer. For the first time to our knowledge, transient oscillations were observed in DPS and the spectral shift of a probe pulse was time resolved together with the rise in DPS, which is clear evidence for induced phase modulation in absorptive materials.

The study of femtosecond absorption spectroscopy made great progress during the past decade. The pump–probe technique with a white-light continuum pulse has been playing major roles in this progress because difference transmission spectra (DTS) can be obtained by using a multichannel spectrometer without scanning a probe wavelength. On the other hand, femtosecond phase spectroscopy, which is complementary to femtosecond absorption spectroscopy, is still in the primitive stage. This is mainly because there have been no methods by which difference phase spectra (DPS) can be obtained with a multichannel spectrometer as in the case of DTS.

To obtain a phase change on excitation,[1] various time-resolved interferometers have been developed.[2–6] They utilize spatial interference fringes, which require the time coincidence of the reference and the probe pulses from the same light source within the coherence time. To obtain DPS by using this interference, one needs a tunable laser or spectral filtering of a continuum. However, such a measurement is difficult because of the complicated experimental setup needed.

In this Letter we propose a new interferometer, a frequency-domain interferometer, which utilizes interference fringes in the frequency domain. By analogy with the beat, which is interference in the time domain caused by two frequency components, two components in the time domain (the reference and probe pulses) interfere in the frequency domain, as readily derived by the Fourier transform. Spectral interference (frequency-domain interference) can therefore be observed even when the two pulses are displaced by more than the pulse duration, in contrast to spatial interference. This method enables us to obtain DPS and DTS simultaneously with a multichannel spectrometer. How this method works is explained as follows.

Let us consider two identical pulses displaced temporally by T. One pulse is a probe pulse, and the other is a reference pulse. They can be generally expressed by

$$E_{\mathrm{pr}}(t) = E(t)\exp(i\omega_0 t),$$

$$E_{\mathrm{ref}}(t) = E(t - T)\exp[i\omega_0(t - T)],$$

where $E(t)$ is an arbitrary complex function. It is not necessary that $E(t)$ be a real function. Then the Fourier transform of the two pulses is

$$F[E_{\mathrm{pr}}(t) + E_{\mathrm{ref}}(t)] = E(\omega - \omega_0)[1 + \exp(-i\omega T)], \quad (1)$$

where $F[E(t)] = E(\omega)$. Experimentally the time-dependent signals can be Fourier transformed by a grating in a spectrometer and detected as power spectra. The intensity of Eq. (1) is given by

$$|F[E_{\mathrm{pr}}(t) + E_{\mathrm{ref}}(t)]|^2 = |E(\omega - \omega_0)|^2(2 + 2\cos\omega T),$$

which represents the frequency-domain interference with the period of $2\pi/T$.

A general expression of the electric field of a probe pulse propagating in a medium is

$$E_{\mathrm{pr}}'(t) = \frac{1}{2\pi}\int d\omega E(\omega - \omega_0) \times \exp\{i\omega[t - n(\omega)x/c + ik(\omega)x/c]\}, \quad (2)$$

where x is the thickness of the medium, c is the velocity of light, and $n_c(\omega) = n(\omega) - ik(\omega)$ is the complex refractive index of the medium. The Fourier transform of Eq. (2) is obtained as follows:

$$E_{\mathrm{pr}}'(\omega) = F[E_{\mathrm{pr}}'(t)] = E(\omega - \omega_0)\exp\{[-in(\omega) - k(\omega)]\omega x/c\}.$$

Suppose both the reference and the probe pulses are transmitted through the same medium and the probe pulse alone undergoes a change in the complex refractive index, $\Delta n_c(\omega,\tau) = \Delta n(\omega,\tau) - i\Delta k(\omega,\tau)$, owing to the pump pulse, which precedes the probe pulse by τ such that

$$E_{\mathrm{pr}}'(\omega,\tau) = F[E_{\mathrm{pr}}'(t,\tau)] = E_{\mathrm{pr}}'(\omega)\exp\{[-i\Delta n(\omega,\tau) - \Delta k(\omega,\tau)]\omega x/c\}.$$

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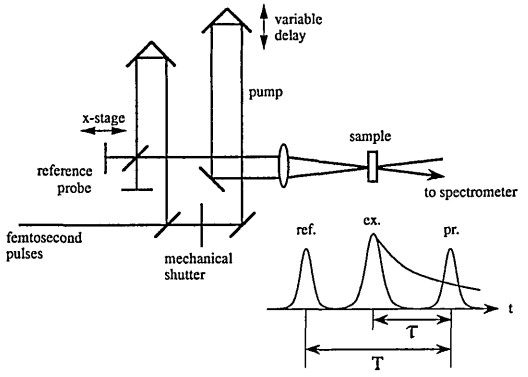

Fig. 1. Experimental setup of the frequency-domain interferometer and the time sequence of the pump (ex.), probe (pr.), and reference (ref.) pulses with separation τ and T.

Then the interference signal without excitation is expressed by

$$|E_{pr}'(\omega) + E_{ref}'(\omega)|^2 = |E_{pr}'(\omega)|^2(2 + 2\cos \omega T), \quad (3)$$

and that with excitation is expressed by

$$|E_{pr}'(\omega,\tau) + E_{ref}'(\omega)|^2 = |E_{pr}'(\omega)|^2\{1 + \exp[-2\Delta k(\omega,\tau)\omega x/c] + 2\exp[-\Delta k(\omega,\tau)\omega x/c] \times \cos \omega[T - \Delta n(\omega,\tau)x/c]\}. \quad (4)$$

Comparing Eq. (4) with Eq. (3), we can obtain simultaneously $\Delta n(\omega,\tau)$ and $\Delta k(\omega,\tau)$ from the peak shifts and amplitude changes of the fringes, respectively.

Temporally separated pulses can interfere owing to the linear dispersion of a grating, which broadens the pulse widths to make them overlap temporally, as explained by Scherer *et al.*[7] When light is incident normally upon a grating, it is diffracted at an angle θ, and optical path differences are made between different transverse components of the light. The components diffracted from neighboring grating grooves interfere constructively if θ satisfies $d \sin \theta = m\lambda$, where d is the period of the grating grooves, λ is the light wavelength, and m is an integer. In the case of a short light pulse, this mechanism causes temporal broadening of the pulse that amounts to $D_0 \sin \theta/c$, where D_0 is the transverse dimension of the pulse. In the case of two pulses displaced temporally by T, the components from the two pulses transversely separated by D can interfere with each other when $D \sin \theta = cT$. Since $D \gg d$, this interference causes a shorter period of modulation as a function of θ, which is frequency-domain interference, than does the interference by $d \sin \theta = m\lambda$.

The experiments were performed by the following procedure. The output of a homemade colliding-pulse mode-locked ring dye laser is amplified by a six-pass amplifier pumped by a copper-vapor laser.[8] The wavelength, duration, energy, and repetition rate of the amplified pulses are 620 nm, 60 fs, 2 μJ, and 10 kHz, respectively. Figure 1 shows the frequency-domain interferometer apparatus, where the time-division technique of Ref. 5 is employed for separating the reference from the probe, but the configuration is much simpler because there is no need for overlapping the pulses temporally. The pulse is divided into two pulses, the pump and the probe. The probe pulse is further divided into two arms of a Michelson interferometer for the reference and probe pulses, and they are displaced temporally by T adjusting one of the arm lengths. The reference and probe pulses then propagate through the same path and are focused into a sample. Both transmitted pulses are detected by a spectrometer with a multichannel photodiode. The pump passes through a variable-delay line and is focused into the sample at a small angle from the reference and probe beams. The pump is blocked at 5 Hz by a mechanical shutter to get signals with and without excitation alternately. Difference spectra are obtained as a function of the time delay τ of the probe from the pump. The reference pulse arrives at the sample earlier than both the pump and probe pulses such that the reference is not affected by the pump. The displacement T was fixed at 410 fs throughout the measurements. The polarizations of all the pulses were parallel, and all the data were taken at room temperature. By blocking the reference beam, the ordinary pump–probe measurement was also performed to obtain DTS.

The setup in Fig. 1 is obtained by adding only two more optical parts, a beam splitter and a mirror, to the pump–probe setup. This is the simplest configuration of all the time-resolved interferometers developed so far and is more stable than a standard two-arm interferometer against several fluctuations because the path difference between the reference and the probe is only the two short arms of the Michelson interferometer.

To demonstrate the method, we measured a commercially available Toshiba R63 glass filter contain-

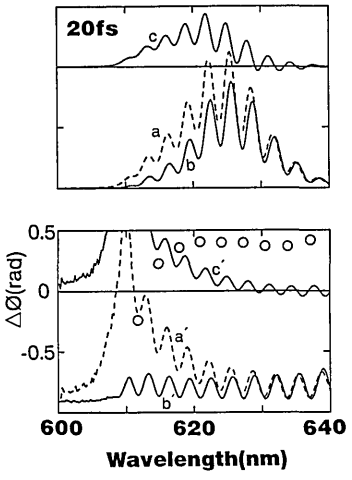

Fig. 2. Signals for the R63 filter obtained by the frequency-domain interferometer at τ = 20 fs and T = 410 fs. Upper panel: directly observed interference spectra with excitation (curve a) and without excitation (curve b); the difference spectrum (curve c) is also shown. Lower panel: curves a, b, and c have been normalized by the transmitted probe spectrum to obtain a′, b′, and c′, respectively. The open circles (DPS) are calculated from the fringe-valley shifts between curves a′ and b′.

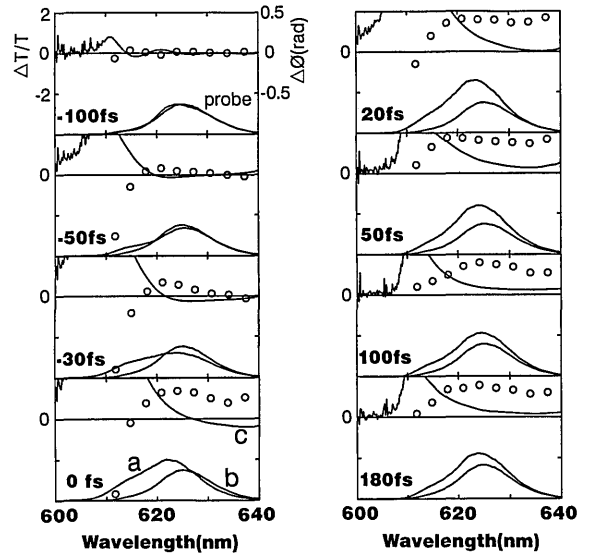

Fig. 3. Transmitted probe spectra with excitation (curve a) and without excitation (curve b) and the DTS (curve c) and the DPS (open circles) for the R63 filter.

ing CdS_xSe_{1-x} microcrystallites of a few weight percent because it has a large nonlinearity and is suited for excitation at 620 nm. Figure 2 shows signals observed by the frequency-domain interferometer at $\tau = 20$ fs and $T = 410$ fs. The excitation density was approximately 3.8×10^{-3} J/cm^2. The DPS are given by the open circles, which were calculated from the ith fringe-valley wavelengths with and without excitation, λ_i^{ex} and λ_i, as $2\pi(\lambda_i - \lambda_i^{ex})/(\lambda_{i+1} - \lambda_i)$. The DPS are averages over 20 cycles of pump on and off taken for 4 s.

To calculate phase shifts, we did not use fringe peaks but fringe valleys because the systematic error that is due to the amplitude change ($\Delta k' = \Delta k \omega x/c$) is estimated as

$$\delta\Delta\Phi \approx \frac{-[\exp(-\Delta k') \pm 1]\mathrm{d}\Delta k'/\mathrm{d}\omega}{T + \mathrm{d}\Delta\Phi/\mathrm{d}\omega},$$

where $\Delta\Phi = -\Delta n \omega x/c$, $\delta\Delta\Phi = \Delta\Phi$ (true) $- \Delta\Phi$ (measured), and the plus and minus denote the fringe peak and valley, respectively. Since $\exp(-\Delta k') > 0$, the fringe valleys cause smaller errors. Both $\exp(-\Delta k')$ and $\mathrm{d}\Delta k'/\mathrm{d}\omega$ are large near 610 nm, where the errors are estimated to be 0.1 rad at most. Except for in the immediate vicinity of 610 nm, however, the errors are negligible compared with the obtained phase changes. Note that this error can be reduced as T is increased.

Figure 3 shows DTS, DPS, and the transmitted probe spectra with and without excitation for several time delays. The DPS show a positive phase change (negative refractive-index change) in average and the sign of the DPS changes at the absorption saturation peak, as expected from the Kramers–Kronig relations.[9] At −100 fs there are oscillatory structures in both DTS and DPS. These are known as transient oscillations,[10] which were observed in DPS for the first time to our knowledge. From −50 to 0 fs, the transmitted probe spectra are blue shifted on excitation. The rapid rise in the DPS from negative to zero delay evidences a major role of induced phase modulation[11,12] in the shift. Without the DPS, we cannot tell whether the transmission change is due to the real absorption change or to the spectral shift of the probe.

Owing to the simple setup, the frequency-domain interferometer can be readily implemented by using a femtosecond white-light continuum[13] to obtain DPS over the whole visible region of the spectrum, and thereby it will open a new field, femtosecond phase spectroscopy.

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