

Phase-sensitive interferometry with ultrashort optical pulses

Rik H. J. Kop and Rudolf Sprik

van der Waals-Zeeman Laboratorium, Universiteit van Amsterdam, Valckenierstraat 65,
1018 XE Amsterdam, The Netherlands

(Received 2 May 1995; accepted for publication 5 September 1995)

Reliable phase-sensitive time-resolved interferometry with ultrashort pulsed lasers is performed with the use of a closed scanning Michelson interferometer in combination with a fixed Mach-Zehnder interferometer at the front end. The technique is based on measuring the full phase and frequency properties of the pulse distortion of an ultrashort optical pulse introduced by linear or nonlinear interaction with a sample. The necessary stability and reproducibility to perform an interferometric measurement is provided by a commercially available Fourier transform spectrometer enabling time-resolved measurements from the IR well into the visible part of the optical spectrum. The feasibility of the technique is demonstrated by measuring the distortion introduced by an etalon and a surface-plasmon polariton. © 1995 American Institute of Physics.

I. INTRODUCTION

We report a method for performing phase-sensitive time-resolved interferometry, using a commercially available Fourier transform spectrometer operating from the IR up to the UV part of the spectrum in combination with a fixed Mach-Zehnder (MZ) at the front end. The technique provides a reliable spectroscopic tool for performing time-resolved interferometry over a wide spectral range. The approach is similar to the method described in Refs. 1–5, and is, in principle, identical to the method already used by Michelson.⁶ All these methods are based on measuring the interferometric cross-correlation signal between an undistorted and a distorted light field. After Michelson, the method reported here was again of interest in Fourier transform infrared spectroscopy.⁷ Recent interest in the method is in the field of ultrashort pulse generation,^{1–5} due to the role of precise pulse evaluation of both intensity and chirp after they have traveled through optical components.

From the cross-correlation signal, both absorption and dispersion in the sample are measured simultaneously, enabling the determination of the group velocity dispersion in materials, for example. In particular, for the distortion of an ultrashort optical laser pulse, the chirp and the sign of the chirp of the pulse distortion can be obtained in a straightforward way without any assumptions.

In a pump-probe type of setup, the time resolution is limited by the time duration of the pulses. The shortest reliable time scale in this type of experiment is the time duration of the pulses. However, in our reported method no such limitation occurs, but the shortest reliable time scale is in the order of the duration of one cycle of the light field used in the experiments.

In the experiment a pulse from a laser is sent through a fixed-length MZ interferometer (Fig. 1) producing two pulses with a separation ΔL . The sample is placed in one of the arms of the MZ interferometer and this laser pulse is distorted by the sample. The pulse in the other arm remains unaltered and will be used as a reference pulse. The electric field of the reference pulse with light frequency ω , may in general be described in the frequency domain by $A(\omega)\exp(i\omega t)$, with $A(\omega)$ being a complex slowly varying

amplitude function. The complex amplitude function $A(\omega)$ describes both the power spectrum of the reference pulse and the relative phase relation between the frequency components. The electric fields of the sum of the distorted pulse and the reference pulse is given by

$$E(t) = \int_{-\infty}^{+\infty} d\omega A(\omega) e^{i\omega t} + \int_{-\infty}^{+\infty} d\omega H(\omega) A(\omega) e^{i\omega(t+\Delta L/c)}, \quad (1)$$

where $\Delta L/c$ is the time delay between the two pulses introduced by the MZ interferometer. The first integral on the right-hand side describes the undistorted pulse, the second the distorted pulse. The distortion of the pulse induced by the sample is described by the transfer function $H(\omega)$. In general, $H(\omega)$ is a complex function of ω , describing both the amplitude of the frequency components and the relative phase relation between the frequency components of the pulse distortion. Using the relative phase relation, the chirp on the pulse can be calculated.

The two pulses are sent into a closed scanning Michelson interferometer, and we measure the interferogram, which is essentially the autocorrelation signal of the double pulse. The signal $I(\tau)$ observed by the detector after exposure to the Michelson interferometer is

$$\begin{aligned} I(\tau) &= \int_{-\infty}^{+\infty} dt E(t) E^*(t-\tau) \\ &= \int_{-\infty}^{+\infty} d\omega H(\omega) |A(\omega)|^2 e^{i\omega(\tau-\Delta L/c)} \\ &\quad + \int_{-\infty}^{+\infty} d\omega (|A(\omega)|^2 + |H(\omega)A(\omega)|^2) e^{i\omega\tau} \\ &\quad + \int_{-\infty}^{+\infty} d\omega H(\omega)^* |A(\omega)|^2 e^{i\omega(\tau+\Delta L/c)}, \end{aligned} \quad (2)$$

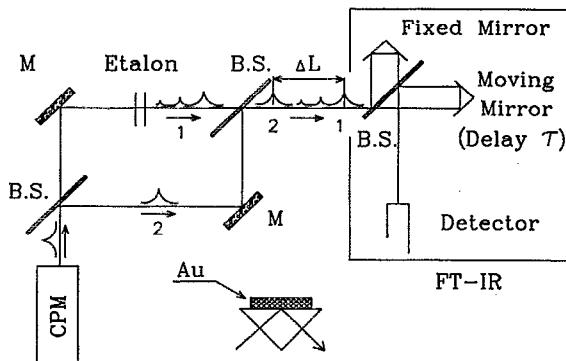


FIG. 1. The experimental setup for time-resolved measuring of pulse distortion. It consists of a fixed Mach-Zehnder interferometer and a scanning Michelson interferometer. CPM: colliding-pulse mode-locked dye laser, BS: beam splitter, M: mirror, FTIR: the Biorad FTS 60A. The inset shows the Kretschmann geometry for excitation of the SPP.

where τ is the delay induced by the scanning Michelson interferometer. Since this is essentially an autocorrelation function of the double pulse, the signal should be symmetrical around the center $\tau=0$.

In our calculation of the field autocorrelation function, we omit a constant background. The full calculation gives a superposition of a constant background (the dc component) given by the mean intensity of the light field and a field autocorrelation function (the ac component). In the experiments however, we only look at the ac component of the detector, thus the field autocorrelation function gives the correct result.

In Fig. 2, which shows a schematic result of a measurement, the response of the detector is indicated as a function of time delay (τ) in the Michelson interferometer. The interferogram contains three peaks. The first and last integrals on the right-hand side of Eq. (2) describe the cross-correlation functions between the altered and unaltered pulse and vice versa. These integrals correspond to the left- and right-most peaks in Fig. 2. The second integral on the right-hand side describes the sum of the autocorrelation functions of the al-

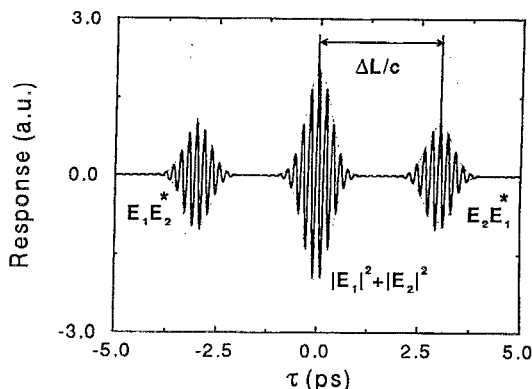


FIG. 2. A schematic plot of the response of the detector as function of time delay (τ) in the Michelson interferometer. The distance between the center of a cross-correlation trace and the center of the autocorrelation trace is given by the time delay between the two pulses ($\Delta L/c$) introduced by the fixed MZ interferometer.

tered and the unaltered pulse, which corresponds to the central peak in Fig. 2. The Fourier transform of one of the cross-correlation functions is

$$\tilde{I}(\omega) = H(\omega) |A(\omega)|^2 e^{i\omega \Delta L/c}. \quad (3)$$

Apart from the constant factor $\exp(i\omega \Delta L/c)$, Eq. (3) is the product of the amplitude function of the reference pulse $A(\omega)$ and the transfer function $H(\omega)$ which describes the pulse distortion. Once $A(\omega)$ is known by analyzing an undisturbed pulse, the full transfer function of the sample can be reconstructed. This is similar to the normal procedure of obtaining, for example, a transmission experiment by the Fourier transform technique. In a transmission experiment, two measurements are required in order to obtain the wavelength-dependent transmission of a sample. However, no direct information is obtained about the dispersion of the sample. Our technique, based on a cross-correlation method, obtains both the absorption and dispersion introduced by the sample in one measurement.

The separation ΔL introduced by the MZ interferometer results in the factor $\exp(i\omega \Delta L/c)$ in Eq. (3). This factor introduces an apparent additional relative phase relation which depends linearly on the frequency. For an accurate determination of the relative phase relation induced by the sample, we have to know the center of the cross-correlation function which defines ΔL . The observed ΔL can be used to determine the group velocity by measuring the difference in ΔL with and without sample. The difference in distance between the center of the interferogram and the center of the cross-correlation function of both measurements is a measure for the change in the optical path length. From this difference and the length of the sample, we can determine the induced group velocity and group index of refraction.

For very intense laser pulses, when nonlinear optical effects start to play a role, a straightforward Fourier analysis breaks down. A more elaborate analysis of the pulse distortion is then required, which should include a model to describe the nonlinear part of the pulse dynamics. In the arguments that have previously been presented, ultrashort optical pulses have been employed with a duration much shorter than the time difference induced by the MZ. The same arguments also apply to incoherent light sources.^{3,4} For our method it is necessary that the coherence length of the incoherent light source be shorter than ΔL . This is necessary because only then is it possible to distinguish between the cross-correlation functions and the central autocorrelation function.

II. EXPERIMENTS AND RESULTS

The experimental setup to measure the interferogram of a double pulse is depicted in Fig. 1. A colliding-pulse mode-locked (CPM) laser producing pulses with a duration of 60 fs at 630 nm is used as a light source. The day-to-day average value of the bandwidth-time interval product varied between 1.0 and 1.3, which results in a bandwidth of approximately 20 nm. The pulses from this CPM laser are sent into a MZ interferometer to create a double pulse. Both pulses are sent into a scanning Michelson interferometer and we measure the interferogram. As a Michelson interferometer, we use a

Biorad FTS 60 A Fourier transform IR spectrometer.⁸ The Michelson interferometer in this spectrometer is sealed, therefore we cannot put the sample in the Michelson interferometer and simply measure the cross-correlation trace between an undistorted and distorted pulse. In the interferometer one mirror is fixed and the other travels over a maximum distance of 10 cm. A He-Ne laser beam which also passes through the interferometer is used to measure the traveled path length of the moving mirror and is also used for dynamic alignment of the interferometer. In our measurements we require subwavelength accuracy to measure reliable results. This is possible because of the dynamic alignment of the spectrometer. Another advantage of the dynamic alignment is the reproducibility of the measurements, which makes it possible to average over several scans. The resolution in our setup is completely determined by the commercially available spectrometer.

In normal operation the spectrometer is used with an internal light source to perform, for example, absorption and transmission spectroscopy in the IR and visible parts of the spectrum. In order to measure an interferometric autocorrelation function, the internal source is disabled and pulsed light is coupled into the spectrometer through an emission port. To know the spectral content of the pulses, the interferometric autocorrelation function of an undistorted CPM pulse is measured with a resolution of 32 cm^{-1} . Data points are measured at every 90 nm mirror displacement, which corresponds to 4 points per He-Ne fringe of the reference laser. For a measurement an average of 256 scans is obtained.

A typical result of an interferometric autocorrelation function of an undistorted CPM pulse is shown in Fig. 3(a). In this figure the response of the detector is plotted as function of the delay in the Michelson interferometer between the pulse and its copy. The inset shows a small part of the interferogram and illustrates the optical fringes in the interferogram. The points in Fig. 3(a) are the measured data points, and the solid line is a guide to the eye. The Fourier transform of the pulse is shown in Fig. 3(b), where we plotted the response as a function of wave number.

Because the transmission characteristics of an etalon is a well-known problem that is completely understood,⁹ we checked the accuracy of the method by measuring the relative phase relation on a pulse introduced by an etalon. The etalon was placed in one of the arms in the MZ interferometer, and the light from the MZ interferometer was sent into the spectrometer through the emission port. Again we measured the interferogram, but now with a resolution of 8 cm^{-1} .

A typical result of the autocorrelation function of the double pulse with one of the pulses chirped by the etalon is shown in Fig. 4(a). In this figure we see the three-peak structure mentioned earlier. The left- and right-hand peaks are the cross-correlation functions between the chirped pulse and the undistorted pulse. The central peak is the sum of the autocorrelation functions of the chirped pulse and the undistorted pulse.

In Fig. 4(b) we show the Fourier transform of one of the cross-correlation functions (the long dashed line). If we look

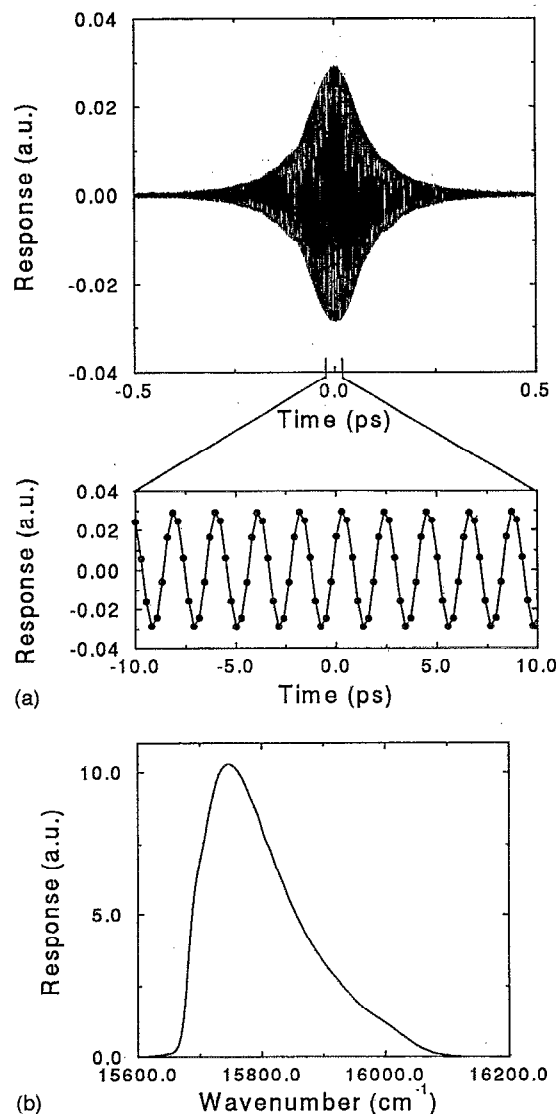


FIG. 3. A typical result of a measurement of the autocorrelation function of an undistorted pulse (a). An enlarged part of the measured autocorrelation function is plotted, showing the resolved fringes in the sampled autocorrelation function. The dots in this figure correspond to data points sampled by the interferometer, the solid line is a guide to the eye. (b) The spectrum of the pulse, which is the Fourier transform of the interferometric autocorrelation function.

at the spectral content of the cross-correlation function and compare this to Fig. 3, we see a sharply peaked spectrum. This is a manifestation of the wavelength-dependent transmission through the etalon. We can extract the separation between the reflecting surfaces in the etalon from the separation between the copies of the pulse in the cross-correlation function to be $73 \pm 5 \mu\text{m}$. From the ratio of the height of the peaks, we estimate the reflection coefficient of one reflecting surface in the etalon to be $r = 0.6 \pm 0.1$. These values are in agreement with those from an independent transmission measurement.

The circles in Fig. 4(b) represent the argument of the complex Fourier transform of the right-hand cross-correlation function of Fig. 4(a). This argument is the relative phase relation between the different components within the pulse or in other words the chirp, as shown in Sec. I. The

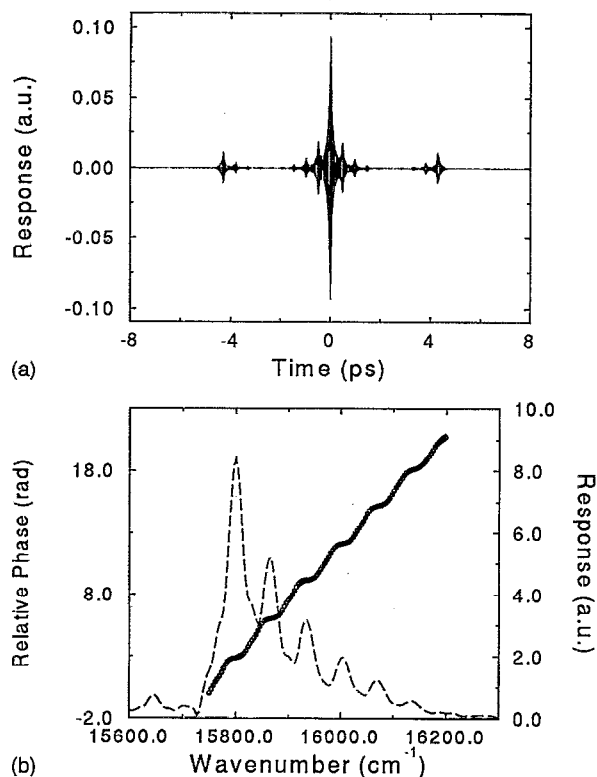


FIG. 4. (a) The measured autocorrelation function of the double pulse with an etalon as a dispersive medium. (b) The spectrum of the right-hand cross-correlation function indicated by the long dashed line (right axis). The solid line in this figure is the theoretical prediction of the relative phase (left axis). The circles are the measured data.

time direction of the relative phase relation on the pulse was determined by keeping track of which path in the MZ interferometer is longer.

The solid line in Fig. 4(b) is the best numerical fit of the argument of the amplitude transmission coefficient calculated with the values found earlier for the spacing between and the reflection of the surfaces of the etalon. This argument gives the relative phase relation between the frequency components. The center of the cross-correlation function (ΔL) was determined by calculating the geometrical "center of mass" of the cross-correlation function with respect to the center of the interferogram. In the fit procedure, we used two parameters. One parameter is an arbitrary offset for the relative phase relation. The second is a correction to the slope which comes from the uncertainty in ΔL . If we compare the experimental result with the result obtained from theory, we see an excellent agreement. We estimate the accuracy of the method from the mean deviation of the difference between theory and experiment to be 0.15 rad.

In a second experiment one of the mirrors was replaced by a prism made of BK7 glass with a thin gold film with a thickness of approximately 32 nm on the hypotenuse face (see the inset of Fig. 1). If the light in the MZ interferometer is p polarized, we can resonantly excite a surface plasmon polariton (SPP) in the gold film by tuning the angle of incidence with respect to the hypotenuse face of the prism. In this way we match the wave vector of the light parallel to the

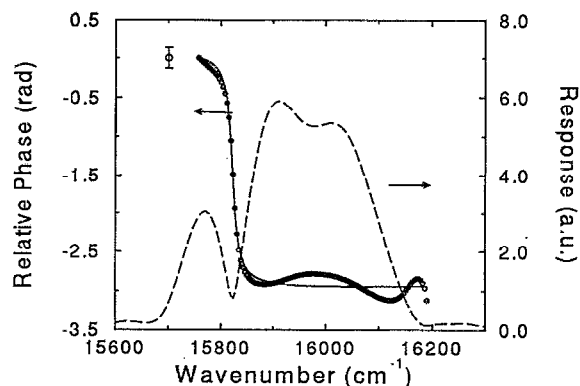


FIG. 5. The spectrum of a phase-sensitive interferometric measurement of the chirp introduced by a SPP indicated by the long-dashed line in the right axis. The measured relative phase relation is indicated with the circles (left axis) and the solid line is a theoretical prediction of the relative phase relation.

hypotenuse and the wave vector of the SPP. This is the so-called Kretschmann geometry.¹⁰

The result of a measurement done with the SPP in one of the arms in the MZ interferometer is shown in Fig. 5. The long dashed line in this figure is the Fourier transform of one of the measured cross-correlation functions. Again we see a sharply peaked spectrum. The circles in Fig. 5 are the data points of the measured relative phase relation of the pulse. The amplitude reflection coefficient from the Kretschmann system can be calculated in a way similar to the transmission through an etalon. Details about this calculation can be found in Ref. 10. The solid line in Fig. 5 is the best numerical fit of the argument of the reflectivity. The same fit procedure is used as in the experiments with the etalon. The parameters found in the calculation of the reflectivity are the same as those used in Ref. 11 for a gold film with a thickness of 42 nm ($\epsilon_{\text{prism}}=2.28$, $\epsilon_{\text{gold}}=-13+1.8i$, $\theta=43.76^\circ$). The error bar in Fig. 5 is the uncertainty found from the experiments done with the etalon. If we compare the experiment with theory we see that the experimentally found relative phase relation is in agreement with theory within the experimental errors.

III. DISCUSSION

We have introduced a novel method, based on a commercially available Fourier transform spectrometer, to perform phase-sensitive time-resolved interferometry. The method is similar to the method introduced by Naganuma.³⁻⁵ With this method it is possible to measure the spectrum and the detailed phase dependence, introduced by an absorbing and dispersive sample. This is demonstrated using ultrashort laser pulses in the visible range of the spectrum.

An advantage of the method is the simplicity of the experimental setup. The dynamic alignment of the spectrometer makes the method highly reproducible and enables averaging over many scans.

In the examples presented of phase-sensitive interferometry, not all the possibilities of the method are used. For instance, it is also possible to do time-resolved measure-

ments. The time resolution in such a measurement is not limited by the time duration of the pulse, but by the time resolution of the spectrometer. In this way it is possible to do measurements with much higher time resolution than obtainable with pump-probe-based measurements.

ACKNOWLEDGMENTS

The authors would like to thank Willem Vos for helpful discussions and comments on the manuscript. This work is part of the research program of the "Stichting voor Fundamenteel Onderzoek der Materie," which is financially supported by the "Nederlandse Organisatie voor Wetenschappelijk Onderzoek."

- ¹W. H. Knox, N. M. Pearson, K. D. Li, and C. A. Hirlimann, *Opt. Lett.* **13**, 574 (1988).
- ²M. Beck and I. A. Walmsley, *Opt. Lett.* **15**, 492 (1990).
- ³K. Naganuma, K. Mogi, and H. Yamada, *Opt. Lett.* **15**, 393 (1990).
- ⁴K. Naganuma and Y. Sakai, *Opt. Lett.* **19**, 487 (1994).
- ⁵K. Naganuma, *Appl. Phys. Lett.* **64**, 261 (1994).
- ⁶A. A. Michelson, *Light Waves and Their Uses* (University of Chicago Press, Chicago, 1902).
- ⁷J. Chamberlain, *Infrared Phys.* **11**, 25 (1971).
- ⁸Bio-Rad, Digilab Division, 237 Putnam Avenue, Cambridge, MA 02139.
- ⁹This can be found in many textbooks, see for instance M. Born and E. Wolf, *Principles of Optics*, 6th ed. (Pergamon, Oxford, 1986).
- ¹⁰F. Kretschmann, *Z. Phys.* **241**, 313 (1971).
- ¹¹Rogier H. M. Groeneveld, R. Sprik, and A. Lagendijk, *Phys. Rev. B* (to be published).