

# Ultrafast heterodyne-detected transient-grating spectroscopy using diffractive optics

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We present a method for implementing optical heterodyne detection using a diffractive optic for time-resolved transient-grating experiments. This technique does not require active phase locking of pulse pairs to achieve interferometric stability. The phase stability, intrinsic time resolution, and signal amplification are demonstrated experimentally through Raman scattering in carbon disulfide. © 1998 Optical Society of America [S0740-3224(98)02505-3]

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A common experimental problem in nonlinear optical spectroscopy is that of detecting extremely weak signal beams with good signal-to-noise ratios (SNR's). At the same time, it is often of physical interest for the detected signal to vary linearly with the material polarization created in the sample under study, rather than quadratically as it will if the signal field is incident on a square-law detector. Optical heterodyne detection (OHD) is a well-established method for achieving both signal amplification and linearization.<sup>1</sup> In OHD, a large steady-state reference field  $E_{\text{ref}}$  is mixed coherently on a detector with the weak time-varying signal field of interest  $E_s(t)$ , so that the total intensity on the detector is

$$I = |E_{\text{ref}}|^2 + |E_s(t)|^2 + 2|E_{\text{ref}} E_s(t)| \cos(\phi_{\text{ref}} - \phi_s), \quad (1)$$

where  $\phi_{\text{ref}}$  and  $\phi_s$  are the phases of the reference and signal fields. The constant background term  $|E_{\text{ref}}|^2$  can be easily subtracted with either a dc filter or through lock-in detection, and in the limit  $|E_{\text{ref}}| \gg |E_s(t)|$  the term quadratic in  $E_s(t)$  can be neglected. This leaves the interference (heterodyne) term, which is linearly proportional to  $|E_s(t)|$  and amplified by a factor of  $2|E_{\text{ref}}/E_s(t)|$  over direct detection.

Previous applications of OHD to nonlinear spectroscopic experiments have shown amplification factors and improvements in the SNR of many orders of magnitude over direct detection.<sup>2</sup> By varying the phase of the reference field, the full complex signal field can be measured rather than simply the magnitude, which often yields greater insight into the physical mechanisms in the sample that give rise to the radiated light. The primary difficulty in implementing OHD experimentally is controlling the relative phase between the reference and the signal,  $\Delta\phi \equiv \phi_{\text{ref}} - \phi_s$ . If this phase is allowed to vary randomly over the time scale of data collection, the experimental signal will be distorted. A widely used solu-

tion to this problem is to actively lock  $\Delta\phi$  using the output of an interferometer as the error signal for a feedback loop that controls the position of one of the optics in the input beam paths.<sup>3-5</sup> While this approach has been shown to work reasonably well, it involves added experimental complexity and cost over direct detection. Alternatively, OHD has been demonstrated with optical setups that have very good mechanical stability, so that the phase drift between pairs of input beams is low over the time scale of data collection.<sup>6</sup> This approach normally requires great care in the mechanical design of the experimental setup, temperature and humidity stabilization, and protection from air currents to achieve interferometric stability at optical wavelengths.

We implemented OHD using a custom diffractive-optic (DO) beam splitter (National Optics Institute) to achieve passive stabilization of  $\Delta\phi$  without the need for either active feedback loops or unusual mechanical stability. A schematic of the experimental apparatus is shown in Fig. 1 for a transient-grating (TG) geometry. This particular geometry is similar to that proposed in Ref. 7, in which a diffraction grating was used to generate pulse pairs for direct-detected cw TG experiments. The two input beams are focused to the desired spot sizes on the DO, which splits each into two (+1 and -1 diffractive orders) with >30% efficiency in each order. The resulting four beams are achromatically imaged onto the sample by a spherical mirror (SM) placed 1 radius away from the sample and DO. The SM is tilted slightly off-axis to separate the incoming and outgoing beams. A half-wave plate controls the amplitude of the reference beam on the detector, and a window of appropriate thickness is inserted in the probe beam to overlap the probe and the reference pulses temporally. A pair of variably tilted microscope cover slips of thickness  $\sim 200 \mu\text{m}$  in the probe beam path provide fine control over the  $t = 0$  position and relative phase of the probe without significantly deviating the

beam in the transverse dimension. The two pump beams intersect at an angle in the sample, forming a grating interference pattern from which the probe is diffracted in the direction of the reference beam. This geometry automatically ensures proper phase matching for any combination of input frequencies<sup>7</sup> as well as spatial and temporal overlap of the signal and the reference pulses on the detector.

The mechanical constraints on the interferometric stability of the phase difference  $\Delta\phi$  are greatly relaxed in this scheme. For the TG experiment,  $\Delta\phi$  can be written as

$$\Delta\phi = \frac{\pi}{2} + \phi_\chi + (\phi_1 - \phi_2) - (\phi_{\text{ref}} - \phi_{\text{pr}}), \quad (2)$$

where  $\phi_1$ ,  $\phi_2$ ,  $\phi_{\text{ref}}$ , and  $\phi_{\text{pr}}$  are the pump, reference, and probe phases respectively,  $\phi_\chi$  is the phase shift owing to the (generally) complex nonlinear susceptibility, and the term  $\pi/2$  is the phase shift between the source polarization and the radiated signal field. Misalignment of any of the DO, the SM, or the sample cell will change the relative path lengths between the two pump beams and between the probe and the reference by the same amount owing to the geometric symmetry of the TG phase-matching condition. Thus the phase shifts between the beam pairs that arise from these misalignments will cancel in the expression for the heterodyne signal phase,  $\Delta\phi$ . This property eliminates phase noise that would normally be present owing to mechanical vibrations or relaxations of the optics. Misalignment of any of the individual transmissive elements in the beam paths will of course change the relative phase since only one beam experiences a change in path length; however, the mechanical tolerances required for good phase stability are well within the capabilities of common laboratory mounts. In addition, since the beams are propagating reasonably close to one another (within a few centimeters), changes in optical path length owing to air currents are approximately the same for all four beams and thus do not greatly affect the phase stability.

The utility of a DO in implementing OHD is illustrated in Fig. 2, which is a comparison of time-resolved traces of the diffracted signal from carbon disulfide ( $\text{CS}_2$ ) with the reference beam blocked (direct detection) and unblocked

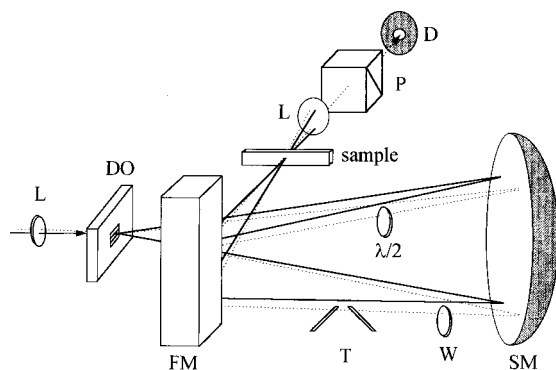


Fig. 1. Schematic of transient-grating experiment using a diffractive optic (DO). The dashed line represents the probe/reference beams and the solid line the pump beams. SM, spherical mirror; FM, folding mirror; T, tilt plate;  $\lambda/2$ , half-wave plate; W, window; L, lens; P, polarizer; D, detector.

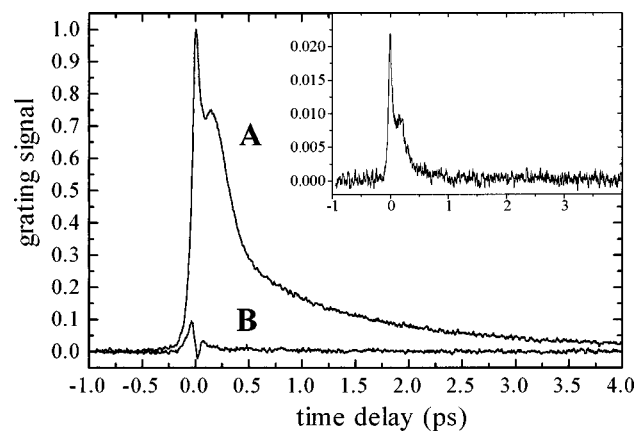


Fig. 2. Transient-grating signals in  $\text{CS}_2$ : (A) OHD signal for  $\Delta\phi = 0$ ; (B) OHD signal for  $\Delta\phi = \pi/2$ . Inset: directly detected grating signal with the reference beam blocked. All signals are normalized to the peak of trace A for reference.

(OHD). The laser source used to collect these data was a regeneratively amplified Ti:sapphire that emitted  $\sim 810$ -nm, 65-fs pulses at 1 kHz. The pump beam was chopped before the DO, and the signal was obtained by lock-in detection to eliminate the reference background  $|E_{\text{ref}}|^2$ . The optimum reference intensity for maximum SNR<sup>8</sup> was chosen to be  $\sim 10^3$  times the signal intensity and was set by rotating the  $\lambda/2$  plate in the reference beam. The distinctive real (Re) and imaginary (Im) signal components, which have been well characterized by others using Kerr-effect spectroscopy,<sup>9,10</sup> were obtained by tilting the cover slip in the probe beam to give a  $\pi/2$  phase shift between collecting the two data sets. These traces correspond to the real and imaginary parts of the  $\chi_{1111}^{(3)}$  susceptibility tensor element probed by this spectroscopy, which are not separable by direct detection.

It should be noted that in order to unambiguously measure the real and the imaginary components of the nonlinear susceptibility, the phase difference  $(\phi_1 - \phi_2) + (\phi_{\text{ref}} - \phi_{\text{pr}})$  between the input fields must be set independently. Since the only experimental observable is  $\Delta\phi$ , which includes the phase shift from the nonlinear susceptibility, it is a nontrivial problem to unambiguously set the field phases at the sample position. This is an issue for any OHD experiment, whether actively or passively phase stabilized, in which none of the input pulses are co-propagating, so that their path lengths to the sample and thus their relative phases are not precisely known. One possible solution to this is to first probe a sample material with a well-known susceptibility (e.g.,  $\text{CS}_2$ ) and then replace it with the sample of interest without perturbing the optical alignment. For the TG experiment on  $\text{CS}_2$ , the Re and Im components of the nonlinear susceptibility were assigned based on the theoretical prediction that the Re signal is proportional to the convolution of the pulse autocorrelation with an effective response function (the susceptibility) and that the Im signal is proportional to the convolution of the derivative of the autocorrelation with the response function.<sup>9</sup> Furthermore, since  $\text{CS}_2$  is almost completely transparent at 800 nm, the Im signal (absorption) is expected to be at least an order of magnitude smaller in amplitude from the Re, so that  $\Delta\phi = 0$ ,

$\pi/2$  approximately corresponds to the real and the imaginary components of  $\chi_{1111}^{(3)}$ , respectively.

To demonstrate the long-time interferometric stability of this setup, we fixed the pump-probe delay to overlap all four input beams temporally and monitored the fluctuations of the diffracted OHD signal from  $\text{CS}_2$  as a function of real time. In order to isolate the phase jitter  $\delta\phi(t)$  between pulse pairs from the noise  $\delta I_L(t)$  that is due to fluctuations in the laser intensity  $I_L$ , the tilt plate in the probe beam was adjusted to set  $\Delta\phi = \pi/2$ . Assuming both phase and amplitude fluctuations to be much less than 100%, the diffracted OHD signal into the detector can be written as

$$S_{\text{OHD}}(t) = [I_L + \delta I_L(t)]^2 \cos\left[\frac{\pi}{2} + \delta\phi(t)\right],$$

$$\approx I_L^2 \delta\phi(t). \quad (3)$$

The measured phase drift, shown in Figs. 3(a) and 3(b) for two different time scales and measurement bandwidths, is about  $\pm 5^\circ$  peak-to-peak ( $2.5^\circ$  rms) over a 10-h period [Fig. 3(b)] and somewhat smaller on a typical data collection time scale of minutes [Fig. 3(a)]. It is notable that this degree of stability is achieved without any attempt to mechanically stabilize the setup. The mechanics involve standard mounts fixed to an optical table and open to the air. For comparison, a Michelson interferometer constructed from similar mounts and physically located near the DO exhibited multiple waves of phase drift under the same environmental conditions.

Owing to the large spectral content of ultrashort pulses, angular dispersion introduced upon transmission through a DO is significant. This does not greatly reduce the time resolution that can be obtained, however, since the angularly dispersed frequencies are focused by the SM to the same spatiotemporal point in the sample. While the signal and the reference output beams will both exhibit angular dispersion following the sample, this does not reduce either the spatial coherence or the time resolution since the dispersion is identical for the two beams and the pulse width following the sample is irrelevant. The capability of this system to achieve high time resolu-

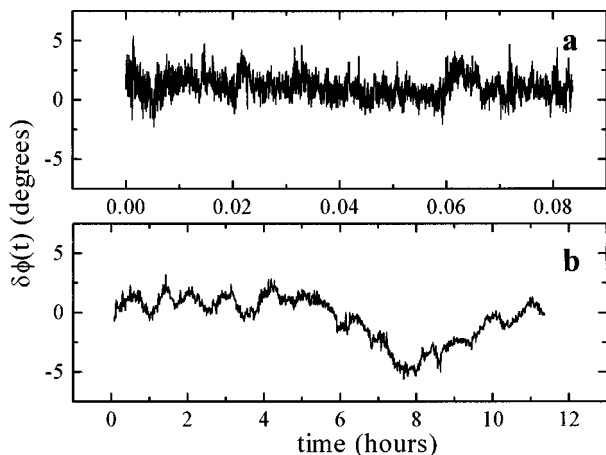


Fig. 3. Time dependence of the phase difference  $\delta\phi$  between input fields: (a) a typical few minutes sampling of  $\delta\phi$  with a bandwidth of 5 Hz; (b) slow drift of  $\delta\phi$  with restricted bandwidth of 0.01 Hz to clarify the long time-scale fluctuations.

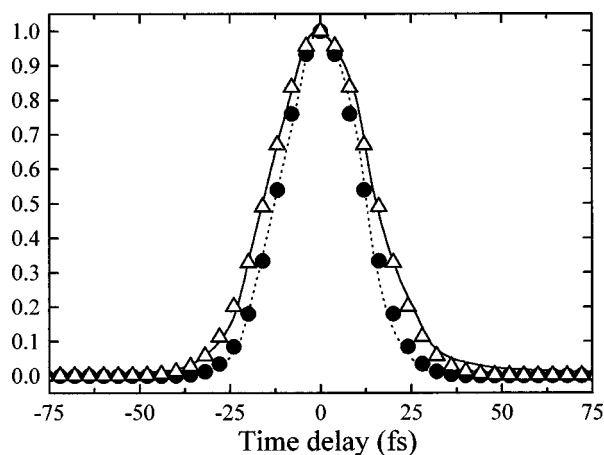


Fig. 4. Comparison of the cross correlation of the undiffracted 0-order pump and probe pulses (dashed curve, FWHM = 25 fs) with the cross correlation between the 0-order probe and +1-order pump (solid curve, FWHM = 32 fs). The symbols are calculated autocorrelation traces for 18-fs intensity FWHM, 35- $\mu\text{m}$ -diameter Gaussian pulses with crossing angles of  $2^\circ$  (circles) and  $15.5^\circ$  (triangles).

tion was probed with  $\sim 18$ -fs pulses directly from a Kerr-lens mode-locked Ti:sapphire oscillator.<sup>11</sup> The undiffracted 0-order pulse was mixed with a +1-order pulse in a second-harmonic-generating crystal (200- $\mu\text{m}$ -thick lithium triborate) to yield the temporal intensity cross correlation, which is then compared with the 0-order autocorrelation as a measure of the true pulse width at the sample. It can be seen from Fig. 4 that the +1/0-order cross correlation is broadened by 7 fs with respect to the 0/0 order correlation. Based on numerical calculations of the autocorrelation envelopes assuming undistorted Gaussian pulses in space and time for the two cases, this broadening can be entirely attributed to geometric smearing of the time delay that arises from the large  $15.5^\circ$  crossing angle and the 35- $\mu\text{m}$  beam diameter at the crystal. This result demonstrates that the DO does not limit the time resolution achievable with this technique beyond that which can be obtained in similar geometries with conventional optics. In addition to the application of diffractive optics on ultrafast timescales, we have also achieved stable heterodyne detection for cw-probed nonlinear grating studies of protein motions that require time scales of milliseconds and longer to characterize.<sup>12</sup> Thus the use of diffractive optics for mixing a signal field with a reference is completely general with respect to dynamic range.

In summary, we have demonstrated optical heterodyne detection on ultrafast time scales in a transient-grating geometry using a diffractive-optic beam splitter to achieve passive phase stabilization without the need for active feedback control of the phase or for exceptionally mechanically stable mounts. We note that this method is generally applicable to any nonlinear spectroscopic experiment in which the driving fields for the nonlinear polarization consist of temporally and spectrally overlapped pulse pairs. Thus, in addition to transient-grating experiments, any nonresonant Raman-scattering experiment can in principle be performed, either with a single custom DO or with several separate DO beam splitters

and appropriate imaging optics. This method may enable simple and robust implementation of OHD in higher-order nonlinear spectroscopies for which active phase controls become prohibitively complex.

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