Double-slit time diffraction at optical frequencies

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Double-slit experiments—where a wave is transmitted through a thin double aperture in space—have confirmed the wave–particle duality of quantum objects, such as single photons, electrons, neutrons, atoms and large molecules. Yet, the temporal counterpart of Young’s double-slit experiment—a wave interacting with a double temporal modulation of an interface—remains elusive. Here we report such a time-domain version of the classic Young’s double-slit experiment: a beam of light twice gated in time produces an interference in the frequency spectrum. The ‘time slits’, narrow enough to produce diffraction at optical frequencies, are generated from the optical excitation of a thin film of indium tin oxide near its epsilon-near-zero point. The separation between time slits determines the period of oscillations in the frequency spectrum, whereas the decay of fringe visibility in frequency reveals the shape of the time slits. Surprisingly, many more oscillations are visible than expected from existing theory, implying a rise time that approaches an optical cycle. This result enables the further exploration of time-varying physics, towards the spectral synthesis of waves and applications such as signal processing and neuromorphic computation.

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Wave–matter interaction in time-varying media exhibits strikingly different dynamics than in conventional passive media. As energy can be exchanged between the wave and medium, the wave frequency changes, and the propagation is no longer symmetric under an inversion of time. Temporal diffraction of matter waves was predicted by Moshinsky in 1952\textsuperscript{1} and has been observed as a few oscillations in the particle arrival time\textsuperscript{2–4}. Amidst the recent explosion of studies on optical time-varying media\textsuperscript{5}, time refraction\textsuperscript{6–8} and time reversal\textsuperscript{9} have been achieved in epsilon-near-zero (ENZ) semiconductors, for example, indium tin oxide (ITO)\textsuperscript{10,11}, modulated by ultrafast laser pulses.

In this Article, we report the observation of the temporal analogue of a double-slit experiment for light waves, showing a clear signature of spectral oscillations for time-diffracted light, and an inversely proportional relation between slit separation and period of oscillations. Moreover, the observed oscillations serve as a sensitive probe of the ITO response time, which we measure to be of the order of an optical cycle, much faster than previously thought.

In the conventional Young’s double-slit experiment, the diffracted pattern has a characteristic oscillatory profile, with minima corresponding to momenta $k$ for which destructive interference suppresses wave propagation (Fig. 1a). These minima have a separation in momentum space that increases for decreasing spatial separation of the slits. In the Fraunhofer approximation of diffraction, that is, for aperture size much smaller than the observation distance, the field distribution in the far field can be approximated as the Fourier transform $\tilde{A}(k_x)$ of the aperture function $A(x)$ (Fig. 1b). The far-field interference pattern can be plotted in a dispersion diagram, where it is described as horizontal transitions (Fig. 1c), with new modes appearing, characterized...
Direct evidence of time diffraction from the temporal double slit is given by illuminating the sample with a probe pulse (230.2 THz carrier frequency and 1.0 THz bandwidth) of duration 794 fs (FWHM) and by monitoring the reflected probe spectrum. The probe pulse is spectrally broadened, exhibiting new frequency content up to 10 bandwidths away from the carrier frequency. A clear spectral modulation, with sinusoidal oscillations, is evident in Fig. 2a,b (red lines), shown for two different time-slit separations. Measurements with 800 fs slit temporal separation (Fig. 2a) present much faster oscillations than with 500 fs (Fig. 2b). The separation of the time slits determines the period of oscillations in the frequency spectrum, whereas the shape of each time slit constrains the oscillation decay and therefore the number of oscillations that are visible. Up to six oscillations in the spectrum are evident (Fig. 2a,b), with an overall decay in intensity away from the central frequency of the probe.

These spectral oscillations are very well captured by a simple diffraction model (Fig. 2a,b, purple line). This model calculates the spectral evolution of the probe from the Fourier transform of the product of the time-varying reflection coefficient $r(t)$, that is, the slit aperture function, and the probe-pulse temporal profile. In such a model, the slit separation and decay time of each slit are fixed by the pump–probe data (Extended Data Fig. 1), whereas the rise time is taken as a fitting parameter.

The oscillation decay with increasing change in frequency depends on the shape of the time slits, particularly their rise time. Therefore, the measured spectral oscillations act as a very precise measurement of the ITO response time, with a resolution well beyond that of a conventional pump–probe experiment. Here, we find unexpected physics: many more oscillations are visible than expected from existing theory, implying a rise time for the leading edge estimated to be 1.0–10.0 fs, that is, of the order of an optical cycle (4.4 fs); this is much faster than previously thought, although recent work has shown the evidence of a speeding up of the response time for pump intensities beyond the linear regime. Moreover, the oscillations are very close to the asymptotic limit of an ideal time slit with a Heaviside (infinitely fast) rise profile, for which the amplitude of the oscillations is expected to decay as $1/(f_f - f_r)^2$, where $f_r$ is the frequency of oscillation and $f_f$ is the probe central frequency (230.2 THz). This is evident when the spectrum is rescaled by the square of the inverse frequency, as its intensity is almost constant (Fig. 2c,d). The dashed purple line in Fig. 2c,d is the asymptotic theory with two Heaviside time slits (Extended Data Fig. 2).

The signature of time diffraction is further revealed by the period of these oscillations, which is inversely proportional to slit separation (Fig. 2e). The agreement between the measured data (open circles) and model is remarkable. The error bars show the accuracy of the slit separation (50 fs) and period (<0.15 THz). The shaded area identifies time-slit separations smaller than 300 fs where the two time slits start to merge and the oscillations are not visible anymore.

Further insight into the temporal diffraction process can be achieved by analysing the full interferogram of the time-diffracted light as a function of slit separation (Fig. 2f). Oscillations appear more pronounced on the red side of the spectrum with frequencies as far out as 10 THz (~60 nm) and only exhibiting frequencies <4 THz away on the blue side (on top of the probe-pulse initial spectral content of 1 THz width). The asymmetric interferogram is explained by the time evolution of the phase of the complex reflection coefficient, causing a Doppler shift in the spectrum, often dubbed time refraction. The theoretical plot (Fig. 2g) does not capture this asymmetry, as it does not include a phase change during refraction. The observed spectral redshift is reproduced when using a dispersive material-based model that includes a time modulation of the phase of the reflection coefficient (Extended Data Figure 3c).

**Outlook**

In conclusion, we report a direct observation of spectral oscillations—at optical frequencies—resulting from double-slit time diffraction, the
The temporal analogue of Young’s slit experiment. The measurements show clear inverse proportionality between the oscillation period and time-slit separation. These oscillations reveal 1–10 fs temporal dynamics of the ITO/gold bilayer, much faster than previously thought and beyond the adiabatic and linear intensity dependence assumed so far in most theoretical models but compatible with recent modelling, calling for a new fundamental understanding of such ultrafast non-equilibrium responses. The observation of temporal Young’s double-slit diffraction paves the way for the optical realizations of time-varying metamaterials, promising enhanced wave functionalities such as non-reciprocity, new forms of gain, time reversal and optical Floquet topology. The visibility of oscillations can be used to measure the phase coherence of the wave interacting with it, similar to wave–matter interferometers. Double-slit time diffraction could be extended to other wave domains, for example, matter waves, optomechanics, electronics, and spintronics, with applications for pulse shaping, signal processing and neuromorphic computation.

Online content
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Methods

Temporal and spectral characterization of probe and pump pulses

All the pulses are generated by a PHAROS (Light Conversion) solid-state laser, set to 230.2 THz with a Light Conversion’s ORPHEUS optical parametric amplifier, with a nominal FWHM of the electric-field envelope of 225 fs. The pulses are roughly Gaussian in shape.

For the double-slit experiment, a probe pulse identical to the pump pulse is sent through a 4f system, composed of a diffraction grating, variable aperture and two lenses, which separates, filters and recombines different spectral components to broaden the pulse in time, to make it longer than the time-slit separation. By selecting the spectral content of the probe beam with an aperture (Extended Data Fig. 3d (pink line); the original spectrum in beige is given for reference), the pulse is extended in time. This is quantified by using cross-correlation measurements, where we collect the sum-frequency-generated signal originating from the spatial overlap of the probe and pump pulses in a 400-µm-thick gallium phosphide crystal with second-order nonlinearity.

As the analytical solution to the cross-correlation measurement is known for a Gaussian pulse, we fit the FWHM of the probe-pulse amplitude assuming such a shape following the measurement of the sum frequency generated between the pump and probe pulses (Supplementary Fig. 1). The agreement between theory and experiment shows that this is a well-suited approximation for the main peak of the probe envelope. For reference, the pristine pump pulse autocorrelation measurement and fit are also measured. The probe pulse is measured to have 794 ± 37 fs FWHM, whereas the original pump pulse is recorded for it from this measurement, and instead, the spectral data (Fig. 2) are necessary.

From this measurement, we can also set the relative amplitude of the second peak of the time slit to be 0.93% of the first one. As the rise time is limited by the probe pulse, we cannot estimate a precise value for it from this measurement, and instead, the spectral data (Fig. 2) are necessary.

Our model effectively reproduces the rise and decay time of modulation but does not account for the short plateau at the maximum of modulation, which arises when pumping near or beyond the saturation of the mirror’s temporal response. As time-varying effects exclusively depend on the slope of the modulation, we focus on reproducing this behaviour rather than the extent of the flat region at the maximum.

A wide range of separations can be achieved by varying the delay between the two pumps (Extended Data Fig. 1b, horizontal axis), whereas the delay of the probe is varied to map the modulation (Extended Data Fig. 1b, vertical axis).

An intensity dependence study of the modulation is illustrated in Extended Data Fig. 1c, showing that the double-slit experiments are performed where the modulation contrast of the time-varying mirror is saturated with pump power. The two pumps have a similar effect on the reflectivity change of the sample, as it assumes the same temporal response at all frequencies. It also neglects the time evolution of the phase of \( r(t) \), which is, in principle, a complex function but is assumed to be real here.

Although this model effectively reproduces the period and amplitude of the oscillations of the double-slit experiment, it fails to capture the asymmetry observed in the experimental spectrum, where more oscillations are visible on the red side (comparing Extended Data Fig. 3a, b). We attribute this to the time evolution of the phase of the complex reflection coefficient, which causes a Doppler shift of the double-slit experiment, the same illumination and detection apparatus is used, with the exception that the probe is temporally broadened before the sample by passing through an additional 4f system. We refer to our previous work for a complete characterization of the ITO/gold sample’s linear and nonlinear properties.

The double-slit aperture function (reflectivity \( R(t) = |r(t)|^2 \), where \( r(t) \) is the complex Fresnel coefficient) can be measured by pump–probe spectroscopy (Extended Data Fig. 1a, blue line), where the probe reflectivity is plotted as a function of delay for a fixed slit separation. The theoretical fit (Extended Data Fig. 1a, dashed red line) comes from the amplitude reflection coefficient \( r(t) \) defined earlier, convoluted with the envelope of the probe pulse in time \( E_{\text{probe}}(t-r) \) and pulse arriving at delay \( t \) (approximated to a Gaussian profile with 225 fs FWHM), which is why spectrally integrated measurements cannot determine the reflectivity rise time.

In addition to fixing the values of normalization constants \( A, B \) and \( C \), the fitting of the pump–probe reflectivity measurements allows us to assign a value for the \( \beta \) coefficient at 1/400 fs\(^{-1} \), corresponding to a decay time of intensity reflectivity (defined as the time to decrease from 90 to 10%) of 625 fs (or 1/e of 330 fs, compatible with the literature\(^5\)).

Modelling of the slits

We model a single-slit aperture function as:

\[
 f_{ss}(t) = \frac{1}{(1 + e^{-\alpha t}) \times (1 + e^{\beta t})}.
\]

where \( \alpha \) and \( \beta \) are taken to be positive constants. The double-slit modulation profile of the amplitude reflection coefficient can then be expressed as \( r(t) = A \times f_{ss}(t - \frac{S}{2}) + B \times f_{ss}(t + \frac{S}{2}) + C \), where \( S \) is the slit separation, \( A \) and \( B \) are the respective slit amplitudes, and \( C \) is a constant corresponding to the unmodulated reflection coefficient of the time-varying mirror. This function is normalized by fitting it to the measured intensity reflectivity change \( R(t) = |r(t)|^2 \) (Extended Data Fig. 2a).

In Fig. 2c, d, we compare the decay of frequency oscillations with an asymptotic case, where the rise time becomes infinitely fast and the decay becomes infinitely slow (that is, when \( \alpha \to \infty \) and \( \beta \to 0 \)). We model this limiting case using two consecutive Heaviside functions (denoted as \( H(t) \)) (Extended Data Fig. 2b) using the following expression:

\[
 r(t) = A \times H(t - \frac{S}{2}) + B \times H(t + \frac{S}{2}) + C.
\]

Characterization of the temporal double slit

To characterize the double-slit time modulation, we use a short 225-fs FWHM probe in a degenerate pump–probe experiment. The probe is incident on the sample at a 60° angle to the normal, whereas the pump beams are incident at a 6° angle difference on either side of the probe. All the beams are \( p \) polarized to couple them to the Berreman resonance. The probe and one of the pumps go through a delay line to control the probe arrival time on the sample and the temporal separation between the two. The reflected probe signal is sent to a Princeton Instruments NIRvana camera for spectral characterization. For the
spectrum, often dubbed as time refraction. A phenomenological model of the phase evolution cannot be built from our measurements, as the pump–probe data are only sensitive to the amplitude of reflectivity.

To better understand this asymmetry, we also model time diffraction using an adiabatic time-varying model that accounts for the phase evolution of the reflection coefficient as well as material dispersion (Extended Data Fig. 3c). Permittivity modulation is computed by calculating a negative shift in the plasma frequency from the convolution of the Gaussian pump intensity with an exponential rise and decay response function. This model of the pump-induced modulation in ENZ materials can reproduce the spectral oscillations, period and interferogram asymmetry (Extended Data Fig. 3a); however, it fails to capture the full spectral extent of the oscillations because of the fast rise time of the experimental slit aperture function, which is fundamentally limited by pump duration in our adiabatic simulations. This discrepancy highlights the need for new models to describe such non-adiabatic modulation.

The diffraction model can be used to fit the experimental spectral data, giving a value for parameter \( \alpha \) of 1/2 \( \text{fs}^{-1} \), corresponding to an intensity reflectivity rise time (10–90%) of 7 fs. As shown in Extended Data Fig. 3d, the data are close to the asymptotic limit of an infinitely fast rising slit; therefore, the fitting is equally good for a reflectivity rise time (10–90%) in the range of 1–10 fs.

**Data availability**

Source data are available for this paper and are available via a public repository at https://doi.org/10.6084/m9.figshare.21968435. All other data that support the plots within this paper and other findings of this study are available from the corresponding authors upon request.

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**Competing interests**

The authors declare no competing interests.

**Additional information**

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Extended Data Fig. 1 | Characterization of the temporal double slit with a short (225 fs) probe pulse. a, Experimental intensity reflectivity (blue line) for a 2.3 ps separation between the time slits, as a function of the probe delay. This is fitted with the model in Fig. S2A (dashed red line). b, Reflectivity for a linear variation of the slit separation (horizontal axis) measured as a function of the probe delay with one of the pumps (vertical axis). c, Intensity dependence of the maximum achievable reflectivity for the two time slits driven by pump 1 (incident at 54°) and pump 2 (incident at 66°). The reflectivity saturates reaching a maximum value of 0.6.
Extended Data Fig. 2 | Modelled reflection coefficient. 

**a**, Amplitude reflection coefficient $r(t)$ corresponding to a realistic modulation of the time-varying mirror (see Fig. S3A). **b**, Amplitude reflection coefficient in the asymptotic limit.
Extended Data Fig. 3 | Comparison of the models against experimental data. 

a–c, Signal against frequency and slit separation for experiment (a), the time diffraction model (b) and the adiabatic, dispersive time-varying model (c). The plots are color-saturated to ensure a fair quantitative comparison between the fringe visibility of the respective datasets. 

d, Experimental oscillation spectrum on a logarithmic scale at a slit separation of 800 fs compared to the theoretical one for various material response times.