A tilted pulse-front setup for femtosecond transient grating spectroscopy in highly non-collinear geometries

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Abstract

We demonstrate a tilted pulse-front transient grating (TG) technique that allows to optimally utilize time resolution as well as TG line density while probing under grazing incidence as typically done in extreme ultraviolet (EUV) or soft x-ray (SXR) experiments. Our optical setup adapts the pulse front tilt of the two pulses that create the TG to the grazing incident pulse. We demonstrate the technique using all 800 nm femtosecond laser pulses for TG generation on a vanadium dioxide film. We probe that grating via diffraction of a third 800 nm pulse. The time resolution of 90 fs is an improvement by a factor of 30 compared to our previous experiments on the same system. The scheme paves the way for EUV and SXR probing of optically induced TGs on any material.

Keywords: transient grating spectroscopy, ultrafast spectroscopy, pulse front matching

(Some figures may appear in colour only in the online journal)

1. Introduction

Transient grating (TG) spectroscopy is a particular form of four wave mixing (FWM) spectroscopy providing time-resolved information about the sample’s third order susceptibility. The grating is formed by two identical laser pulses that hit the sample simultaneously under a certain angle, which translates into a spatial modulation of the excitation. A third pulse is scattered from that grating with variable delay after the excitation. Since its invention, TG spectroscopy has been used on numerous samples \cite{1–3} to elucidate the complex dynamics of molecules and solid materials.

We have recently performed TG spectroscopy studies of the ultrafast insulator-to-metal transition in VO\textsubscript{2} \cite{4, 5}. We employed two 800 nm femtosecond laser pulses to trigger the insulator-to-metal transition via a TG. We probed its temporal evolution by diffracting a broadband extreme ultraviolet (EUV) pulse from high harmonic generation (HHG) off the TG. The use of EUV or soft x-ray (SXR) probe wavelengths in combination with TG experiments is a promising pathway to new insights into charge/energy transfer phenomena, exciton dynamics in strongly correlated systems, and structural dynamics in thin films and surfaces due to the element and site specificity of spectroscopy in this wavelength range. Accordingly, we were able to pin down the insulator-to-metal transition site specifically at the vanadium M-edge in our EUV experiments \cite{4, 5}.

In general, EUV and SXR wavelengths are only efficiently reflected at grazing incidence angle. For their efficient diffraction, on the other hand, high line density TGs must be generated. The latter scales with the angle between the optical excitation pulses and is best achieved by illumination of the sample perpendicular to the bisecting line of this angle. A large disadvantage of this highly non-collinear geometry is, however, a considerable pulse front mismatch between pumps and probe compromising the achievable time resolution. In
of the pump beams or the probe beam. In principle, it would be more straight forward to tilt the pulse front of the probe pulse since this only affects one beam. However, this would be challenging in the case of short wavelength pulses. We therefore employ a scheme to tilt and match the pulse fronts of the optical pump pulses. The pulse front tilt angles ($\alpha^+$ and $\alpha^-$) for the two pump pulses have to satisfy the following non-trivial expression:

$$\alpha^\pm = \arcsin \left( \frac{\cos \theta_{pu}}{B^\pm} \right)$$  \hspace{1cm} (1)

with

$$B^\pm = \sqrt{[\cos \theta_{pr}^\pm \pm \sin \theta_{pu}]^2 + \cos^2 \theta_{pu}}^{1/2}. \hspace{1cm} (2)$$

It is possible to obtain such expression imposing that the three beams, overlapping each other on the sample, uniformly keep the pump-probe time delay constant. This condition, referring to figure 1, is satisfied when $MO = NO = PO$.

The matching condition could be fulfilled by individually tilting the fronts of the two pump pulses. Aiming to reduce the degrees of freedom in the alignment procedure, we employ a more elegant design for the production and recombination of the two pump beams using a phase mask. Our new setup intrinsically matches their pulse fronts employing a combination of a blazed grating (BG in figure 2), which generates the pulse front tilt, with a transmission grating (‘phase mask’—PM in figure 2—Hololor, DS-234-800-Y-A, optimized for $\lambda = 800$ nm, fused silica substrate, 22.5 $\mu$m line period), which separates the two pump pulse arms in its $\pm 1$ diffraction orders. Pulse front matching is ensured by confocal imaging the transmission grating on the sample surface (S) [12].

Combining the grating equation and equation (1), it can be shown that the $\pm 1$ diffraction orders after the PM have the desired pulse-front tilt if the PM is orthogonal to the 800 nm beam direction and the latter has a pulse-front tilt $\beta$ (see figure 2(a)) so that

$$\tan \beta = (\cos \theta_{pr}^\prime)^{-1}. \hspace{1cm} (3)$$

Comparing equations (1) and (3), the simplification introduced with this design is self-evident. We would like to stress the fact that $\beta$ is only a function of the probe incident angle $\theta_{pr}^\prime$ and not of the incidence angles of the pump pulses. This allows for modifying the line spacing of the TG independently of the pulse front tilt by replacing the PM.

To induce a pulse front tilt $\beta$ at the PM position (figure 2(a)), we use a reflective diffraction grating with the characteristic equation

$$\sin \gamma + \sin \delta = Nm\lambda, \hspace{1cm} (4)$$

where $\gamma$, $\delta$, $N$, $m$, and $\lambda$ are the angle between the incoming beam and the normal to the grating surface, the diffraction angle, the line density of the grating, the diffraction order and the wavelength of the scattered light. Here, we use the convention $\delta > 0$ if at the same side of $\gamma$ with respect to the
normal to the grating surface. It is easy to show that the 1st diffraction order is characterized by a pulse-front tilt $\beta$ so that

$$\sin \gamma = \frac{\cos (\delta + \beta)}{\sin \beta}. \quad (5)$$

Using a reflecting BG (figure 2, Thorlabs, $N = 830$ lines mm$^{-1}$), we can satisfy both (3) and (5) with $\delta = 44.26^\circ$ and $\gamma = -1.94^\circ$. In order to compensate for the horizontal angular dispersion due to the diffraction, we use two confocal cylindrical lenses (CL1 and CL2 in figure 2, both $f = 5$ cm) to image the grating on the PM. Imaging the BG surface onto the PM instead of focusing the pump beam into the PM has the additional advantage that intensities are kept well below thresholds for nonlinear effects and damage in the PM. By employing the combination of a cylindrical lens (CL3) and a spherical lens (SL, both $f = 15$ cm), we simultaneously image the PM in the horizontal plane onto the sample surface and focus the pump beams in the vertical plane. The resulting spot shapes are sketched in figure 2(b) for several positions in the beam path from BG to the sample. By using the horizontal imaging/vertical focusing scheme, we ensure efficient pumping of the complete sample area, which is irradiated in grazing incidence by the probe pulse. This area is additionally increased by the fact that the probe pulse intersects with the sample before its focus in the detector plane. The optical setup allows to conveniently alternate the polarizations of pump and probe beams using halfwave plates. To demonstrate the functionality of the optical setup, we perform all 800 nm TG experiments in vertical polarization at room temperature on a 100 nm thick single-crystal VO$_2$ film, which is prepared by pulsed laser deposition on a Al$_2$O$_3$ (100) substrate [13]. The employed fluences are well below the damage threshold ($<20$ mJ cm$^{-2}$) [4]. The probe beam has an estimated fluence of 1 mJ cm$^{-2}$. The delay between excitation and probe pulses is implemented via a computer-controlled mechanical delay stage. The TG signal is detected by a photodiode in first-order diffraction direction of the TG. To avoid contamination of the transient signal, the intense beam in zero-order direction is blocked by a beam stopper (BS). To improve signal-to-noise ratio, we used a combination of a chopper wheel in the pump beam path and a boxcar averager in "toggle"-mode. We additionally correct the signal for laser fluctuations. The transients shown in figure 3 are averaged over 2000 laser pulses per delay step.

3. Results and discussion

Figure 2(c) shows the interference pattern between ±1 order diffraction created from the 800 nm pulses with tilted pulse fronts in the PM, imaged onto a CCD camera. The image clearly shows a modulation in intensity along the horizontal axis. We estimate a groove density of $\sim 100$ lines mm$^{-1}$ ($\sim 10$ $\mu$m pitch), which corresponds to an angle of incidence of $\theta_{pu} = 2.29^\circ$ for the ±1 first-order diffraction in agreement with our angle of incidence determinations. In order to test the effectiveness of the pulse-front tilt approach, we performed TG measurements at room temperature. By rotation of BG and thereby either directing the 1st or 0th diffraction order onto the PM, the effect of pulse front matching can be directly assessed. Figure 3 shows a comparison between transients with and without matched pulse fronts.

In figure 3(a), we show a transient obtained with matched pulse fronts. At zero pump-probe delay, we observe a rapid turn on of transient diffraction intensity followed by a decay on the sub-ps time scale. The transient further evolves in form of a second, slower signal increase reaching a plateau at $\approx 3$ ps. We will use the sub-ps dynamical signature to assess the width of the instrument response function of the experimental setup. We, thus, fit the transient with a trust-region-reflective least squares
algorithm using the following phenomenological function

\[ g(t) = G(t) \otimes H(t)(Ae^{-\frac{t}{\tau_1}} + Be^{+\frac{t}{\tau_2}} + C), \]  

whereas \( H(t) \) is the Heaviside step function, \( \tau_1 \) and \( \tau_2 \) exponential time constants, and \( A, B, \) and \( C \) amplitudes. The function is convoluted with a Gaussian approximating the instrument response function. The fitted time constants are \( \tau_1 = (80 \pm 40) \) fs and \( \tau_2 = (740 \pm 90) \) fs. The fitted variance of the Gaussian instrument response function is \( \sigma = (90 \pm 10) \) fs. This is considerably beyond the instrument response function expected from 30 fs laser pulses, but agrees with our estimations of the pump pulse temporal broadening based on the amount of glass in the pump beam path. In principle, the duration of the pump pulses could be further increased by a spatial chirp from tilting the pulse fronts. However, the spatial chirp can be expected to be virtually zero at the sample position due to imaging of BG onto PM and PM onto the sample surface (see figure 2).

We use the fit results to subtract the signal evolution on the time scale of several hundred fs from the sub 100 fs dynamics, which is shown in the inset of figure 3(a). Comparison with the pure instrument response function, which is shown as a dashed green line, clearly shows a broadening of the feature towards positive pump-probe delays due to ultrafast dynamics in the sample. The underlying dynamics are, therefore, on a similar time scale like the instrument response.

For comparison, we show in figure 3(b) a transient obtained with the 0th diffraction order of the BG i.e. unmatched pulse fronts. It is important to notice the different time scales of the two plots in figures 3(a) and (b). To allow a quick comparison between the two time profiles, we report the fit obtained for the tilted pulse front case in orange in figure 3(b). It is evident that the unmatched pulse fronts prevent observation of any sub-ps dynamics. The signal shape is now clearly dominated by the instrument response function. In order to estimate the time resolution of the unmatched pulse front case, we applied the same fit keeping the time constants fixed at the values obtained for the tilted pulse-front case of figure 3(a). It results in a width of the instrument response function of \( \sigma = (2.8 \pm 0.2) \) ps, in agreement with our earlier results [4]. The fit is shown as a red line in figure 3(b).

It is not the goal of this work to clarify the microscopic origin of the spectral shape shown in figure 3(a). Nevertheless, we can qualitatively compare our results with the ones obtained with ultrafast transient holography experiments available in the literature [14–17]. The technical conditions among our work and the ones cited here are different in many respects, including the optical setups, the crystal orientations, the thickness of the samples, the fluences, the laser wavelength, and the substrates. It is not surprising that there is not a quantitative agreement on the measured time scales in play. Nonetheless, it is very clear that the shapes of the signals are very similar: all the experiments, as in our case, present a sharp peak at time zero (in our case we are able to measure also its decay profile) and a slower rising feature in the sub-ps regime. The first ultrafast feature is assigned to the formation of bipolaronic charge-transfer excitons, while the sub-ps dynamic can be related to the insulator-to-metal transition, possibly mediated by electron–phonon coupling.
Future experiments replacing the 800 nm probe beam with EUV pulses from HHG can be performed in a very similar fashion like our previous experiments [4, 5]. They aim at studying the insulator-to-metal transition by observing the collapse of the band gap between the conduction and valence bands. Since both bands are almost exclusively comprised of vanadium-centered 3d orbitals, the collapse can be selectively observed at the vanadium M-edge (37 eV). Thus, future EUV TG experiments must be able to spatially distinguish contributions from consecutive harmonics of the HHG probe in the 37 eV photon energy range and 1st order diffraction direction. Apart from the improvement in time resolution, our modifications to the original setup, in which we succeeded in distinguishing those harmonics close to the vanadium M-edge, slightly change the grating pitch from 15 μm in the previous experiments to 10 μm. The decrease in grating pitch, however, yields a stronger dispersion of the harmonics. Thus, we can expect to distinguish contributions from individual harmonics in the 37 eV range with our modified setup.

4. Conclusion

We demonstrate in this contribution a versatile optical setup for investigating ultrafast dynamics on surfaces combining highly non-collinear pump-probe geometries with sub 100 fs time resolution. The pulse front mismatch resulting from the non-collinear geometry is compensated by tilting the pulse fronts of the optical pump pulses in an easy-to-align setup. We demonstrate matching of the pulse fronts of pump and probe pulses with all-optical experiments on the insulator-to-metal transition in a thin film of VO₂. The setup is ideally suited for future experiments replacing the optical probe pulse by broadband EUV pulses from HHG.

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