Nonlinear polarization holography of nanoscale iridium films

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Abstract

Attosecond nonlinear polarization spectroscopy designates the subcycle-precise retrieval of the electric field of a femtosecond laser pulse together with the nonlinear polarization response that the laser pulse triggers in a sample. Here, we introduce a method that is all-optical and applicable to metal films. The method is called nonlinear polarization holography because it is based on the comparison of two time-domain holograms with and without a metal film on a substrate. The working principle can be understood as the time-domain analog of holographic interferometry, in which the comparison of two spatial holograms reveals changes in an object’s size and position with interferometric precision (i.e. to fractions of the wavelength). Analogously, nonlinear polarization holography provides subcycle precision (i.e. to fractions of the optical period). Nonlinear polarization holography is used here to retrieve the time-domain nonlinear response of a nanoscale iridium film. Using density matrix calculations it is shown that the knowledge of the nonlinear response with subcycle precision allows distinguishing excitation and relaxation mechanisms of low-energetic electrons.

1. Introduction

The propagation of light through matter is influenced by the polarization with which the material responds to the incoming light fields. While the linear part of the polarization is captured by introducing the refractive index of the material, the treatment of the nonlinear polarization is more complex. The nonlinear polarization response \( P^{(NL)} \) acts as a source term in the wave equation and emits a field that superimposes the original light field. For example, the frequency-degenerate part of \( P^{(NL)} \) leads to nonlinear absorption if the emitted field destructively interferes with the original light field. The time-domain retrieval of both the original light field \( E(t) \) and \( P^{(NL)}(t) \) with subcycle precision is called attosecond nonlinear polarization spectroscopy [1, 2]. In previous measurements using femtosecond laser pulses in dielectrics, \( P^{(NL)} \) is reconstructed from the electric fields of the laser pulses measured by an attosecond streak camera after propagation through the samples. Measurements of \( E(t) \) and \( P^{(NL)}(t) \) with subcycle precision reveal the energy flow between light and matter in time-domain [1].

For metals, where strong interaction between light and conduction band electrons exist, attosecond nonlinear polarization spectroscopy has not yet been demonstrated. More traditional measurements of the nonlinear response, e.g. the z-scan procedure [3], usually describe the nonlinearity with a single value such as the nonlinear refractive index \( n_2 \) or the third-order nonlinear optical susceptibility \( \chi^{(3)} \). As recently analyzed, reported values of \( \chi^{(3)} \) of gold span a range of more than three orders of magnitude [4]. The
reported values depend strongly on the pulse duration and on the measurement procedure. These conflicting results are ascribed to the fact that the frequency-domain description of the third-order nonlinear response generally requires three frequencies, i.e. $\chi^{(3)}(\omega_1, \omega_2, \omega_3)$ [5]. The simplified description by a scalar or by a one-dimensional function $\chi^{(1)}(\omega)$, which underlies the measurement procedures mentioned above, is only meaningful in the limit of monochromatic light or an instantaneous response $P^{(NL)}(t) \propto E(t)^3$. It fails when mechanisms that accumulate during a laser pulse, such as heating or interband transitions of electrons, contribute significantly to the nonlinear response. The wide range of reported values for $\chi^{(3)}$ of gold suggests that this is the case for metals.

Methods for measuring $E(t)$ of femtosecond laser pulses, such as frequency-resolved optical gating (FROG) [6], usually rely on a simple relation of electric field and nonlinear response, such as $P^{(NL)}(t) \propto E(t)^3$, and therefore usually cannot determine both $E(t)$ and $P^{(NL)}(t)$. Recently, it has been shown that the FROG procedure can be extended such that an unknown parameter of the nonlinear response function of a dielectric (the decay time of TiO$_2$ [7]) can be determined. Applications to metals or to more complex relations between $E$ and $P^{(NL)}$ are not yet reported.

All-optical methods of two-dimensional electronic spectroscopy [8] could in principle yield equivalent information as attosecond nonlinear polarization spectroscopy but the so-called phasing problem prevents the time-domain retrieval of $P^{(NL)}(t)$. The phasing problem (time-zero problem in the time domain) describes the problem that the relative time delay between the pulse sequence that triggers the nonlinear response and a reference pulse, called the local oscillator, must be known with subcycle accuracy. Approaches for solving the phasing problem presented so far are based on calibrating the optical beam paths with interferometric precision [9–11] or by post-processing the two-dimensional spectroscopy data using the projection-slice theorem when additionally pump-probe data is available [12]. The disadvantages of these approaches include limited precision and a reduction of signal-to-noise ratio [8]. Time-domain retrievals of $E(t)$ and $P^{(NL)}(t)$ based on two-dimensional electronic spectroscopy are not yet reported.

Here, an all-optical method of attosecond nonlinear polarization spectroscopy is introduced and applied to iridium (Ir) films. The method is a variant of time-domain holography and therefore called nonlinear polarization holography, although the purpose is different from typical applications of time-domain holography. Typically, time-domain holography is used to encode digital data in a probe pulse via the nonlinear interaction with a pump pulse in a sample [13]; this relies on a simple relation between $E(t)$ and $P^{(NL)}(t)$ in the sample. Unlike that, the purpose of the present method is not to store digital data in the probe pulse but to reveal the response function of the sample. To achieve that, two holograms are recorded. The first hologram is recorded using a sample (a dielectric substrate) with a known nonlinear response function. The two holograms is used to retrieve $P^{(NL)}(t)$ of the Ir film. No assumptions about the interaction in the metal film are required; also non-perturbative responses of the metal film can be retrieved.

The method is the time-domain analog of holographic interferometry, in which the comparison of two spatial holograms reveals changes in an object’s size and position with interferometric precision [14]. Similarly here, the comparison of two time-domain holograms reveals $P^{(NL)}(t)$ of the Ir film with subcycle precision. The phasing problem is solved in nonlinear polarization holography because the time delay between the probe and the reference pulse (the local oscillator) is constant for both holograms and truncates the retrieval. The approach is applied here to a pump-probe experiment but is also suitable for two-dimensional nonlinear spectroscopy.

Both instantaneous and cumulative contributions are found in $P^{(NL)}$ of the Ir film. To identify the mechanisms of the instantaneous and cumulative contributions, density matrix calculations are performed. It is found that the data can be reproduced reasonably well using a model with two electronic bands. The instantaneous contribution is ascribed to anharmonic electron motion in the bands. The cumulative contributions are ascribed to electron transitions between the bands and heating of the electron gas. Furthermore, the calculations reveal that the mechanisms of relaxation, such as the collisions and dephasing of electrons and holes, strongly influence the subcycle timing of the nonlinear response.

2. Experimental results

Three near-infrared (NIR) pulses interact in a sample with polarization perpendicular to the plane of incidence in pump-probe geometry (figure 1). The pump pulse $W$ with electric field $W(t)$ (peak intensity $I_W = 100 \text{ GW cm}^{-2}$, beam waist 100 $\mu$m) crosses a beam of two weak pulses (probe $U$ and reference $V$ with fields $U(t)$ and $V(t)$, peak intensity $I_U \approx I_V \approx 1 \text{ GW cm}^{-2}$, beam waist 100 $\mu$m) that have a delay of $\theta \approx 770 \text{ fs}$ with a crossing angle $\alpha = 1.5^\circ$. The delay $\theta$ limits the time window for the reconstruction and should therefore be chosen sufficiently large, but at the same time small enough for the interference fringes to be spectrally resolved. The crossing angle $\alpha$ leads to a temporal smearing of the measurement and should
Nonlinear polarization holography. Nonlinear polarization holography uses a probe wave U and a reference wave V with constant time delay $\theta$. The pump pulse W arrives at a variable delay $\tau$ at the sample, which is a dielectric substrate for the first hologram and additionally an Ir film for the second hologram. The spectrally resolved intensities (the holograms) exhibit an interference pattern. $E_S$ and $E_F$, which are the fields emitted by the nonlinear responses of the substrate and the Ir film, are contained in the second hologram, but only $E_S$ is contained in the first hologram.

Therefore be chosen small, but large enough to be able to separate the beams after the interaction [15]. The intensity of the weak pulses should generally be as low as possible since the method is based on the assumption that there is no nonlinear interaction through U and V only. In practice, however, the intensity of U and V must be sufficient for the measurement statistics. Here, U and V are produced by reflections of W from a 80 µm-thick fused silica plate at near-normal incidence, and $\theta$ equals the delay between the front and rear reflections. The temporal shapes of the pulses are very similar (center wavelength: 800 nm; pulse duration: 28 fs), as they originate from the same laser pulse, but this is not a prerequisite for the functionality of the method; U and V only need to cover the same spectral range. The laser repetition rate is 1 kHz. The recorded data result from pulse delay scans with scanning step 0.23 fs that are repeated 600 times and averaged. The detector integration time is 1 ms so one laser shot per scanning step is recorded. No drifts in the pulse delay scans are observed, which indicates that the sample is in thermal equilibrium or quasi-equilibrium, meaning that the temperature is the same each time the laser pulses arrive. No irreversible changes are observed in the sample; however, a significant increase in pump intensity (by at least a factor of two) rapidly degrades the film, presumably by laser ablation.

For the first hologram, the sample is a 80 µm-thick substrate made of silica. Optical spectra are recorded for varying delay $\tau$ of W. This yields the hologram $I_S(\omega, \tau) = |U(\omega) + V(\omega) + E_S(\omega, \tau)|^2$, where $E_S$ is the electric field emitted by the nonlinear polarization response of the substrate. The scalar treatment of the effect is appropriate in the present case because all fields have the same polarization, and the materials are isotropic. No assumption is made about the generation of the fields emitted by the nonlinear polarization response except that the present analysis is restricted to the fields emitted into the probe direction. In the present experiment, cross-phase modulation (XPM) of the probe by the pump is the dominant mechanism, but in other experimental regimes, cascaded processes involving self-diffraction [15, 16] could contribute without limiting the applicability of nonlinear polarization holography. The field emitted by the linear polarization is omitted in the above expression of $I_S(\omega, \tau)$ for the sake of clearer equations; this field is independent of $\tau$ and therefore truncates in the holographic reconstruction [15].

For the second hologram, the sample is replaced by a substrate with a 5 nm-thick Ir film on the rear side produced by atomic layer deposition (ALD), see appendix. Coatings of Ir are interesting for optical applications because they do not require protective layers due to their high intrinsic stability and have a high reflectivity into x-ray frequencies [17]. In the present experiment, we found that the Ir films withstand higher laser intensities than gold films. The reason is likely related to the high melting point of roughly 2700 K, which also makes Ir well-suited for high-temperature plasmonics [18]. To minimize the influence of thickness and repositioning aberrations, the same substrate is used for both measurements. The substrate is partially covered by the Ir film and shifted perpendicular to the laser beams for the measurements with and without Ir film (figure 1). This yields the hologram $I_{SF}(\omega, \tau) = |U(\omega) + V(\omega) + E_S(\omega, \tau) + E_F(\omega, \tau)|^2 |T(\omega)|^2$. In this expression, the linear polarization of the Ir film is accounted for by its linear intensity transmittance $|T(\omega)|^2$ with the linear amplitude transmittance $t(\omega)$. The originally measured field emitted due to
the nonlinear response of the Ir film incident on the spectrometer is $E_S(t) = E_k(t)$, but, to allow for a comparison on equal footing we generate $E_S$ by linear back-propagation through the Ir film.

The holographic reconstruction is a digital procedure analogous to irradiating a spatial hologram with the reference beam (see appendix). The wave $U$ is uniquely determined by the pulse retrieval using the first hologram except for the absolute phase and time zero [19], which carries on to the reconstruction of $E_S$ and $E_F$. However, the relative phases and times of $U(t)$, $E_S(t)$ and $E_F(t)$ are retrieved from the holograms. This is equivalent to the information provided by attosecond nonlinear polarization spectroscopy using an attosecond streak camera [1], except that the absolute phase is not determined in the present experiment. However, the absolute phase could be determined additionally if the method is used with stabilized carrier-envelope phase. For valid results of the pulse retrieval, the intensity must be sufficiently low to ensure that the interaction in the substrate is in the perturbative regime with an instantaneous nonlinear response. This condition is excellently met in the present case [1]. However, the method makes no such assumption about the interaction in the metal film, meaning that also non-perturbative and non-instantaneous responses of the metal film can be retrieved.

The retrieved nonlinear response, presented in figure 2, shows that a wave is emitted from the nonlinear polarization of the substrate only when $U$ and $W$ overlap temporally, see figures 2(a) and (b). The waveforms of $E_S$ and $U$ are approximately the same, but $E_S$ is temporally shifted with respect to $U$, always towards the pump pulse resulting in a tilted ellipse in figure 2(a). The phase is shifted with respect to $U$ by $-\pi/2$, i.e. the phase-lag between $E_S$ and $U$ is a quarter of an optical cycle. Both the pump-induced delay as well as the phase shift of $E_S$ is consistent with the action of XPM induced by an instantaneous nonlinearity. For monochromatic waves, this is commonly expressed by an intensity-dependent refractive index $n = n_0 + n_2 I$, known as the optical Kerr effect. With macroscopic propagation, focusing (self-focusing due to a Kerr lens) occurs. For a peak intensity of the pump-pulse of $I_W = 100 \text{ GW cm}^{-2}$ this agrees with the literature value for $n_2$ of fused silica ($2.73 \times 10^{-20} \text{ m}^2 \text{ W}^{-1}$ [20]). As our set-up generates reliable results for the well-known nonlinear response of fused silica we can now go on and measure the much less investigated nonlinear response of the Ir film.

The wave $E_F$ is emitted by the nonlinear polarization of the Ir film when the pump $W$ arrives simultaneously with the probe $U$ or earlier (negative delay), indicating a cumulative nonlinearity. The stripe pattern in figure 2(c) is addressed to intensity fluctuations of the laser. They influence the holographic reconstruction as seen from (16). For $\tau < -30 \text{ fs}$ (pump before probe), the waveforms of $E_k$ and $U$ are about the same. This is consistent with the assumption that the earlier-arriving pump changed the properties of the

**Figure 2.** Nonlinear polarization response. The envelope of the wave emitted by the nonlinear polarization response of the substrate is displayed in (a)–(d) show the corresponding quantities of the Ir film. The amplitudes are displayed in (a) and (c), the phase-lags with respect to the probe field $U$ in (b) and (d). Curves are displayed for $\tau = -60 \text{ fs}$ ((e)–(g)), and (h) show the probe $U(t)$ (red) and the pump envelope $X(t)$ normalized to their amplitudes (blue, see appendix). (f) and (i) show the electric fields emitted due to the nonlinear responses; (g) and (j) show their phase-lags with respect to the probe pulse for the substrate (black) and for the Ir film (magenta). $E_S$ is very weak for $\tau = -60 \text{ fs}$ and hence not displayed in (f) and (g). The solid lines are calculated using the full dataset (average of 600 pulse delay scans); the dotted lines in (g) and (j) are calculated using fractions of the same dataset (average of 200 pulse delay scans) to get an insight into the statistics. In all panels, $E_k$ and $E_F$ have been normalized to the maximum of $E_S$. For better comparison, $E_S$ is scaled by 3, because $E_S$ is about three times stronger than $E_F$. The envelopes $f_f, f_f, f_f$ are calculated from the fields according to $E_S(t) = \mathcal{R}\{f_f(t) \exp[i\omega_0 t]\}$ with $\omega_0 = 2.36 \text{ PHZ}$ and $n = S, F, U$. Only regions where the amplitude is greater than 15% of the maximum are shown in (b), (d), (g) and (j).
Ir film, altering its complex refractive index. The phase-lag with respect to \( U \) is less than \( \pi/2 \), effectively amplifying \( U \). In other words, the preceding pump saturates absorption and increases the transmission through the Ir film. This bleaching does not decay within our measurement range of 100 fs. For temporal overlap of probe \( U \) and pump \( W \), further changes occur. We again see a temporal shift of \( E_p \) with respect to \( U \) indicating a partially instantaneous response similar to the substrate. But, overall the waveforms of \( E_p \) and \( U \) are significantly different. The pre-pulse of \( U \) is almost not visible in \( E_p \), whereas the post-pulse is considerably amplified due to the reduced absorption (figure 2(i)). This results in an island that appears in figure 2(c) at \( \tau \approx 0 \) and \( t \approx 50 \) fs. This again indicates a delayed or cumulative response, but now within the duration of the pulse. To sum up we have experimentally found evidence for a mixture of instantaneous and delayed responses in Ir films. We will now shed light on the internal dynamics of these processes through numerical simulations, with a particular focus on fast relaxation phenomena.

3. Density matrix calculations

The density matrix calculations are similar as in [21, 22]. The calculations cover electron motion in \( k \)-space through acceleration by the laser electric field (intraband transitions) and dipole transitions of electrons across bands (interband transitions). The equation of motion of the \( k \)-resolved density matrix is [23, 24]

\[
\frac{\text{d}}{\text{d}t} \rho_{lm}^{k} = -\omega_{nm}^{k} \rho_{lm}^{k} + E \cdot \sum_{l} \left( \mathbf{d}_{lm}^{k} \rho_{nl}^{k} - \mathbf{d}_{nl}^{k} \rho_{lm}^{k} \right) + i E \cdot \frac{\text{d}}{\text{d}t} \rho_{lm}^{k} + i \left( \partial_{\rho_{lm}^{k}} \right)_{\text{relax}}
\]

(atomic units are used). These equations are identical to the semiconductor Bloch equations in one spatial dimension, except that the Coulomb interaction is neglected [25]. The matrix elements \( \rho_{nm}^{k} \) are the populations; \( \rho_{nm}^{k} (n \neq m) \) are the coherences of states \( n \) and \( m \). The transition energies are \( \omega_{nm}^{k} = \omega_{nm} - \omega_{n}' \). The terms proportional to \( \frac{\text{d}}{\text{d}t} \) accelerate the electrons and holes within the bands (intraband transitions). To ease the calculations, the coordinate transform \( k \rightarrow k + A \) is applied to (1), where \( A \) is the vector potential defined by \( E = -\partial_{A} A \):

\[
\frac{\text{d}}{\text{d}t} \rho_{lm}^{k+A} = -\omega_{nm}^{k+A} \rho_{lm}^{k+A} + E \cdot \sum_{l} \left( \mathbf{d}_{lm}^{k+A} \rho_{nl}^{k+A} - \mathbf{d}_{nl}^{k+A} \rho_{lm}^{k+A} \right) + i \left( \partial_{\rho_{lm}^{k+A}} \right)_{\text{relax}}.
\]

The initial band population at \( t = -\infty \) is implemented as the Fermi–Dirac distribution at room temperature (293 K). The calculations are based on one conduction band and optionally one valence band as defined in figure 3. The valence to conduction band transitions \( \mathbf{d}_{12}^{k} = \mathbf{d}_{21}^{k} \) are implemented as [25]

\[
\mathbf{d}_{12}^{k} = \mathbf{d}_{12}^{k=0} \frac{\omega_{12}^{k}}{\omega_{12}}.
\]

with \( \mathbf{d}_{12}^{k=0} = 10 \) au.

Whereas the band structure parameters \( \omega_{nm}^{k} \) and \( \mathbf{d}_{nm}^{k} \) can in principle accurately be calculated by \textit{ab initio} methods like density functional theory, the relaxation term \( \left( \partial_{\rho_{nm}^{k}} \right)_{\text{relax}} \) is usually implemented as a phenomenological damping term and suited to test assumptions about the relaxation mechanism.

Transitions of conduction band electrons to the valence band via spontaneous decay are neglected here because the lifetime (the \( T_{1} \) time) usually exceeds 100 fs [26]. Redistribution in \( k \)-space induced by collisions with collision time \( T_{c} \) is implemented either as

\[
\left( \partial_{\rho_{nm}^{k}} \right)_{\text{relax}} = -\frac{1}{2 T_{c}} \left( \rho_{nm}^{k} - \rho_{nm}^{k} \right)
\]

or

\[
\left( \partial_{\rho_{nm}^{k}} \right)_{\text{relax}} = -\frac{1}{T_{c}} \left( \rho_{nm}^{k} - f_{\text{FD}} \left( \omega_{n}^{k} \right) \frac{\sum_{k} \rho_{nm}^{k} \left( \omega_{n}^{k} \right)}{\sum_{k} f_{\text{FD}} \left( \omega_{n}^{k} \right)} \right).
\]

The approach in (4) follows [27] and attenuates the current by symmetrizing the \( \rho_{nm}^{k} \) distribution. Alternatively, the approach in (5) attenuates the current by decay directly into the equilibrium distribution, where \( f_{\text{FD}} \) is the Fermi–Dirac distribution at room temperature (293 K).

For the coherences,

\[
\left( \partial_{\rho_{nm}^{k}} \right)_{\text{relax}} = -\frac{1}{T_{c}} \rho_{nm}^{k}.
\]
account for the nanoscale structure of the Ir film. The band shapes are approximated by \( \omega = \omega_b + k_b (1 - \cos(k a) - b_1 (\cos(3ka) - \cos(ka))) \) with \( \omega_b = -1.4 \text{ eV} \), \( b_2 = 4 \text{ eV}, b_2 = 2 \) for the conduction band and \( \omega_b = -1.5 \text{ eV}, b_1 = 1.45 \text{ eV}, b_2 = 2 \) for the valence band with the lattice constant \( a = 0.384 \text{ nm} \). The shape of the conduction band is similar as calculations on https://materialsproject.org, the shape of the valence band is chosen to enable interband transitions within the optical spectrum of the laser pulse. The real (imaginary) part of the linear refractive index of the two-band model is calculated with (12) and experimental reference data [17] (black dashed) are shown in (b) and (c). The blue line shows the refractive index solely due to intra-band transitions in the conduction band (calculated with (10)), the green line shows the refractive index solely due to inter-band transitions (calculated with (11)).

\[ T_2 \] is the interband dephasing time. Here \( T_2 = 2T_c \) is assumed for all coherences and independent of \( k \).

The polarization \( P \) and the current \( J \) are calculated by

\[
P = N\epsilon \sum_{n \neq m} \sum_k d_{nm}\rho_{nm}^k \delta k \tag{7}
\]

\[
J = -N\epsilon \sum_k \sum_n n_n^k \delta k, \tag{8}
\]

where \( v_n^k = \partial_k \omega_n^k \) is the electron group velocity and \( \delta k \) is the spacing in the \( k \)-grid. The macroscopic observables are scaled to an effective electron number density \( N_e \), because the two-band approximation in one dimension does not reflect all contributing electrons.

The resistivity of this model is

\[
\rho = \frac{1}{N_e T_c \left(v_r^l - v_r^h \right)}, \tag{9}
\]

where \( v_r^l \) and \( v_r^h \) is the electron group velocity of the conduction band at the right and the left Fermi edge. For the present calculations, \( N_e = 0.0045 \) is chosen which reproduces the literature value \( \rho = 47 \text{ nF/m} = 0.22 \text{ au} \) for \( T_c = 30 \text{ fs} \), which is a typical value for bulk metals [29]. For the Ir film, the collision time \( T_c = 15 \text{ fs} \) is assumed. This value is lower than the value assumed for the bulk material to account for the nanoscale structure of the Ir film [30].

With the linear susceptibility of the intraband transitions

\[
\chi_{\text{intra}}^{(1)}(\omega) = \frac{\mathcal{F}\{J\}}{i\omega \mathcal{F}\{E\}} = N\epsilon \frac{v_r^h - v_r^l}{i\omega T_c - \omega^2} \tag{10}
\]

and the linear susceptibility of the interband transitions

\[
\chi_{\text{inter}}^{(1)}(\omega) = \frac{\mathcal{F}\{P\}}{\mathcal{F}\{E\}} = N\epsilon \sum_k 2\omega_{12}^k \left(\omega_{12}^k\right)^2 \left(\omega_{12}^k\right)^2 - \omega^2 + 1/T_c^2 + 2i\omega/T_c \delta k \tag{11}
\]

the linear refractive index is calculated as depicted in figure 3:

\[
n(\omega) = \sqrt{1 + 4\pi \chi_{\text{inter}}^{(1)}(\omega) + 4\pi \chi_{\text{intra}}^{(1)}(\omega)}. \tag{12}
\]

The numerical calculations are performed on a \( k \)-grid with 37 points. The time-domain integration is performed using the 4th-order Runge–Kutta (RK4) method on a \( t \)-grid with 12 001 points in the interval \([-750, 750] \text{ fs}\). The spacing on the \( \tau \)-grid is 2 fs.

The band structure parameters are displayed in figure 3; the relaxation parameters are summarized in table 1.


| Parameter | Value | Reasoning |
|-----------|-------|-----------|
| $T_1$    | $\infty$ | The $T_1$ time exceeds by far the measurement range of 100 fs [26] |
| $T_c$    | 15 fs | A typical value for bulk metals is 30 fs [29], but collisions in the nanoscale structure of the Ir film are more frequent [30] |
| $T_2$    | 30 fs | The value is not known. The relation $T_2 = 2T_c$ accounts for the fact that an electron collision should affect the interband coherence |
| $N_e$    | 0.0045 | this reproduces the literature value $\rho = 47$ nΩm [28] |

Figure 4. Simulation of the nonlinear polarization response. The same as figure 2, except that synthetic data is displayed using 2 bands, the symmetrizing decay (according to (4)) and constant relaxation times.

4. Discussion

The main purpose of our calculations is to elucidate how the subcycle timing of the nonlinear response is affected by the relaxation mechanisms. Scattering on ions, impurities, grain boundaries, and surfaces as it is particularly pronounced in ALD layers with their island-like structure on the nanoscale is expected to be nearly elastic [30, 31]. Therefore, we implement scattering as a redistribution in $k$-space that attenuates the current but conserves the kinetic energy of the electrons. This is achieved by a relaxation term that symmetrizes the $k$-distribution (see (4) and [27]).

The calculation including one valence band and one conduction band reproduces the data reasonably well (figure 4). For example, the increase of the amplitude of $E_F$ in the region of temporal pulse overlap is qualitatively reproduced. Also, the accumulation of the nonlinearity during the laser pulse, most evident in the form of an island appearing at $\tau \approx 0$ and $t \approx 50$ fs, is consistent with the data (panel (c)). Even the $t-\tau$-dependent phase-lag shown in panel (d) agrees qualitatively within the scanned range of pump-probe delays, only the absolute value of the phase-lag is closer to $-\pi/2$ in the calculation than in the experimental data (panel (j)). This discrepancy between calculation and experimental data is statistically significant (see dotted lines in figure 2(j)). Systematic errors of the measurement that are unknown so far might have an influence. A big influence, however, comes from the model that is used for the calculation, especially the mechanisms of excitation and relaxation, as discussed in the following.

The intraband transitions include an instantaneous (non-cumulative) nonlinearity, which is caused by the anharmonicity of the bands. The contribution of this non-cumulative nonlinearity is revealed by the increase in the amplitude of $E_F$ in the region of temporal pulse overlap. This increase in the amplitude is also reproduced when the calculation includes only the conduction band (see appendix, figure 7), therefore it is ascribed to intraband transitions. The intraband transitions also include a cumulative nonlinearity, which is caused by the redistribution in $k$-space due to the scattering of accelerated electrons towards a non-thermal electron distribution that gradually builds up during the laser pulse (see figure 5(a)). This appears as smearing in $k$-space at the Fermi edge with a width given by the excursion of the electrons, which is proportional to the laser electric field.
A further cumulative nonlinearity is caused by interband transitions [26]. During the laser pulse, electrons gradually appear in the conduction band at \( k \)-positions where the energy gap is in resonance with the laser field (figures 5(b) and 3(a)).

There are further mechanisms of relaxation that are hitherto not included in our calculation. If only the symmetrizing redistribution in \( k \)-space is considered, neither the internal thermalization of the electron gas nor the equilibration of the electronic and lattice temperature (external thermalization) is reached. However, the internal thermalization is expected to be rather slow and comparable with the external thermalization for electrons in the vicinity of the Fermi level [32, 33], in contrast to the fast decay of optically excited electrons due to electron-electron scattering [29]. The comparison of the experimental data with a calculation in which the relaxation term is implemented as a relaxation into the equilibrium distribution (external thermalization) shows that this decay, which does not conserve the kinetic electron energy of the electron gas, is not observed within the time range of this study (see appendix).

Each of the individual mechanisms that cause a nonlinear response in our model (see appendix, figures 7 and 8) affects the subcycle timing of the nonlinear polarization in a characteristic way. Electron motion in anharmonic bands causes a phase-lag of \(-\pi/2\), similar as observed in the fused silica substrate (focusing nonlinearity). Excitation of electrons from valence into conduction bands (interband nonlinearity) causes a phase-lag near \( \pi/2 \) (defocusing nonlinearity). Contrary to that, the symmetrizing decay (intraband nonlinearity) causes a phase-lag near \(-\pi/2\). The contribution of intraband transitions to the linear refractive index (10) suggests that the strength of the intraband nonlinearity depends on the electron group velocity at the Fermi edge and that the phase will deviate from \(-\pi/2\) when the collision time \( T_c \) is very short (comparable to the laser optical cycle). Similarly, the contribution of interband transitions to the refractive index (11) suggests that the strength of the interband nonlinearity depends on the dipole matrix element and that the phase will deviate from \( \pi/2 \) when the dephasing time \( T_2 \) is comparable to the laser optical cycle or the inverse transition frequency. Hence, the \( t-\tau \)-dependent plot of the phase-lag can be regarded as a fingerprint of the relaxation mechanism.

5. Summary and outlook

In summary, nonlinear polarization holography is suited to retrieve the nonlinear response of a nanoscale metal film to an ultrashort laser pulse with femtosecond resolution and subcycle accuracy. Instantaneous and cumulative contributions can be distinguished, and the gradual buildup of the nonlinear response during the laser pulse is observed. The subcycle timing of the nonlinear response with respect to the excitation pulses is particularly valuable for gaining insight into relaxation mechanisms. Different models of relaxation can be tested in calculations of the \( k \)-resolved density matrix. For the case of a nanoscale Ir film, the nonlinear response on the time-scale of 100 fs can be modeled surprisingly well using a rather simple model with two bands. A symmetrizing \( k \)-space relaxation, caused by prevailing near-elastic scattering, proves to influence the ultrafast nonlinear response significantly. A certain disagreement between the phases of experimentally observed and simulated fields hints at a more complex nature of scattering events, which will be investigated in further experiments.
Data availability statement

The data cannot be made publicly available upon publication because they are not available in a format that is sufficiently accessible or reusable by other researchers. The data that support the findings of this study are available upon reasonable request from the authors.

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P P and A S prepared the samples. M H acquired the data. A N P developed the method nonlinear polarization holography, analyzed the data and developed the theory. All authors contributed to the discussion and preparation of the manuscript.

Appendix

A.1. Pulse retrieval

The temporal shapes of the laser pulses are characterized using XPM scans, which is a recently demonstrated method for the simultaneous characterization of two unknown and independent laser pulses [19]. The retrieval method is analytic and generates a retrieved trace. The agreement between the data trace and the retrieved trace is a measure of the fidelity of the pulse retrieval. The traces are complex-valued. Originally demonstrated for the retrieval of two pulses in the deep ultraviolet, which are scanned with a further unknown NIR pulse [34], the method is here applied for three unknown frequency-degenerate pulses U, V, and W using the variant center of the original publication [19].

The basis for the pulse retrieval is the complex trace \( I_2(\omega, \tau) - I_0(\omega, \tau = \infty) \) (see Holographic reconstruction). Fourier transform from \( \tau \) to \( \omega_r \) yields the trace \( Y(\omega, \omega_r) \), which is related to the unknown fields by

\[
Y(\omega, \omega_r) \propto U(\omega + \omega_r) V^*(\omega) X(-\omega_r),
\]

with \( X(\omega_r) = \int d\omega_2 R(\omega_2) R(\omega_r - \omega_2) \). This data trace is the product of three functions that only depend on \( \omega, \omega_r \) and \( \omega + \omega_r \). To ease the numerical retrieval of \( U \) and \( V \), the substitution \( \omega_r \rightarrow -\omega - \omega_r \) is carried out. This results in

\[
Y(\omega, \omega_r) \propto U(-\omega_r) V^*(\omega) X(\omega_r + \omega).
\]

As detailed in [35, 36], the differentiation of the logarithm of the trace with respect to \( \omega \) and \( \omega_r \), followed by the integration of the exponential of the obtained trace along its diagonal provides the preliminary solutions \( \hat{U}(\omega) \) and \( \hat{V}(\omega) \). The relation of the preliminary solutions and the true solutions is \( U(\omega) = U(\omega) e^{i(\alpha + T_\omega + s\omega)} \) and \( V(\omega) = V(\omega) e^{i(\beta + T_\omega + s\omega)} \), with the real integration constants \( \alpha, \beta, T_\omega \) and \( s \). From these expressions, it is apparent that \( T_\omega \), which is the time of arrival, cannot be determined. Also, the simultaneous determination of \( \alpha \) and \( \beta \), which are the carrier envelope phases, is impossible. The integration constant \( s \) can be determined from the experimentally measured spectra \( |U(\omega)|^2 = |V(\omega)|^2 \).

The scanning step (the grid-point spacing of the \( \tau \)-axis) limits the bandwidth in \( \omega_r \) according to the Nyquist–Shannon sampling theorem, which in turn limits the temporal resolution (the grid-point spacing of the \( \tau \)-axis) of the retrieved pulses. The fundamental limit is given by the bandwidth in \( \omega \) (this is the spectral bandwidth of the laser pulses). The scanning step should be chosen such that its Nyquist frequency is higher than the bandwidth in \( \omega \). Laser pulses with broader spectral bandwidth require smaller scanning steps for optimal temporal resolution.

The pulse retrieval from \( Y(\omega, \omega_R) \) is analytic, and the fidelity of the retrieval is checked by comparing the complex-valued data trace with the retrieved trace. The traces and the retrieved pulses are depicted in figure 6.

A.2. Holographic reconstruction

The two holograms \( H_2(\omega, \tau) \) and \( H_2(\omega, \tau) \) are inverse Fourier transformed with respect to the \( \omega \)-axis, the side peak (alternating component) is cut-out and shifted to zero, and thereafter Fourier transformed with the
Fourier transform convention \( F \{ f(t) \} \propto \int_{-\infty}^{+\infty} f(t) e^{-i\omega t} \, dt \). This digital procedure is analogous to the irradiation of a spatial hologram with the reference beam. The shift toward zero in pseudo time should be approximately \( \theta \), but it is not necessary to know \( \theta \) precisely, since it truncates in (15) and (16). This yields the complex-valued traces \( J_{S}(\omega, \tau) = (U(\omega) + E_{S}(\omega, \tau)) V^{*}(\omega)e^{-i\omega \theta} \) and \( J_{SF}(\omega, \tau) = (U(\omega) + E_{S}(\omega, \tau)) V^{*}(\omega)e^{-i\omega \theta} T(\omega) \). The temporal range of \( \tau \) must only encompass the temporal range of \( U \) and \( E_{S} \), not the reference \( V \). Using the complex-valued traces, the fields emitted by the nonlinear responses can be determined:

\[
E_{S}(\omega, \tau) = U(\omega) \left( \frac{J_{S}(\omega, \tau)}{J_{S}(\omega, \tau = \infty)} - 1 \right), \tag{15}
\]

\[
E_{F}(\omega, \tau) = (U(\omega) + E_{S}(\omega, \tau)) \left( \frac{J_{SF}(\omega, \tau)}{J_{S}(\omega, \tau) T(\omega)} - 1 \right). \tag{16}
\]

Here it is assumed that no nonlinear response is induced when the pump arrives after the probe \( (E_{S}(\omega, \tau = \infty) = E_{F}(\omega, \tau = \infty) = 0) \). In the analysis, the averaged data in the range \( \tau > 70 \) fs is used for \( J_{S}(\omega, \tau = \infty) \) and \( J_{F}(\omega, \tau = \infty) \). The transmission is determined by \( T(\omega) = \frac{I_{F}(\omega, \tau = \infty)}{I_{S}(\omega, \tau = \infty)} \). The contribution of \( E_{S} \) in (16) can be neglected in the present case, because \( E_{S} \ll U \) (the biggest value of \( E_{S} \) is found at the fundamental frequency and pulse overlap, where \( \frac{E_{S}(\omega, \theta)}{U(\omega, \theta)} = 0.015 \)).

The wave of \( U \) is uniquely determined by the pulse retrieval except for the absolute phase and time zero, which carries on to \( E_{S} \) and \( E_{F} \) in (15) and (16). However, the relative phases and times of \( U(t) \), \( E_{S}(t) \) and \( E_{F}(t) \) are retrieved from the holograms, similar as the relative phases and times between \( U(t) \), \( V(t) \) and pump envelope \( X(t) \) are uniquely determined by the pulse retrieval.

In the holographic reconstruction, the temporal resolution of \( U \) carries on to \( E_{S} \) and \( E_{F} \). Hence, the bandwidth of the used laser pulses and the scanning steps of \( \tau \) limit the temporal resolution of \( E_{S} \) and \( E_{F} \) according to the Nyquist–Shannon sampling theorem (see Pulse retrieval).

For valid results of the pulse retrieval, the intensity must be sufficiently low to ensure that the interaction in the substrate is in the perturbative regime with an instantaneous nonlinear response, a condition that is excellently met in the present case [1]. However, the method makes no such assumption about the interaction in the metal.
A.3. Comparison of models

![Figure 7](image_url)

Figure 7. Comparison of calculations with 1 band. Calculations including only the conduction band (see figure 3) are displayed with equilibrium decay (according to (5)) or symmetrizing decay (according to (4)). For each calculation, (a)–(j) correspond to the panels in figure 4 in the main paper.

A.4. Macroscopic pulse propagation

Macroscopic pulse propagation is calculated similarly as in [21, 22]. The unidirectional pulse propagation equation (UPPE) [37] is used. The propagation direction $z$ and one transverse dimension (the $x$-dimension) are included; this accounts for the noncollinear geometry. A scalar electric field is used because all pulses are polarized perpendicular to the plane of incidence. The propagation is calculated through the entire sample, which consists of a substrate for the calculation of the first hologram and additionally an Ir film for the calculation of the second hologram. For the substrate, instantaneous nonlinear response is assumed as $P_{\text{NL}}(t) = \chi^{(3)} E(t)^3$ with $\chi^{(3)} = 3.5$ au (that is $1.66 \times 10^{-22}$ m$^2$V$^{-2}$ in SI units [23]). The UPPE reads

$$\partial_z \hat{E} = i \left( \frac{\omega}{u} - \hat{K} \right) \hat{E} - \frac{2\pi \omega}{Kc^2} \left( i \omega P^{(\text{NL})} \right),$$

with $K = \sqrt{n_R^2 \frac{\omega^2}{c^2} - k_z^2}$. Numerical tables are used for the refractive index $n_R$, $c$ is the speed of light and $u$ is the group velocity of the NIR pulse. This includes the linear optical response via the refractive index $n_R$ of the substrate. The hat symbol indicates the Fourier transform in the dimensions of time and transverse space.
Figure 8. Comparison of calculations with 2 bands. The same as figure 7, except that also one valence band (see figure 3) is included. Calculation 2 is identical to the calculation shown in the main text (figure 4 in the main paper).

For the Ir film, both the linear and the nonlinear optical response are contained in the expressions for $P$ and $J$ in (7) and (8) and the UPPE is

$$\partial_t \hat{E} = i \left( \frac{\omega}{\alpha} - K \right) \hat{E} - \frac{2\pi \omega}{Kc^2} (i\omega \hat{P} + \hat{J}), \quad (18)$$

with $K = \sqrt{\frac{\omega^2}{c^2} - k_x^2}$.

The UPPE is integrated numerically using the split-step method with an $x$-grid with 81 points in the interval $[-250, 250] \mu m$ and a $z$-grid with 41 points in the interval $[0, 80] \mu m$ for the propagation through the substrate and 8 points in the interval $[0, 5] nm$ for the propagation through the Ir film.

Subsequent to the propagation inside the sample, the light propagating collinearly to pulses $U$ and $V$ is calculated by

$$E(\omega) = \hat{E}(\omega, k_x) \quad (19)$$

with $\tan\left( -\frac{\alpha}{2} \right) = \frac{k_x}{\sqrt{\omega/c^2 + (k_x)^2}}$.

A.5. ALD

The ultrathin Ir films are prepared on half of the surface area of fused silica substrates by means of ALD technique using a SunALE R-200 ALD reactor (Picosun Oy, Masala, Finland). Iridium(III) acetylacetonate ($\text{Ir(acac)}_3$) and molecular oxygen ($O_2$) as precursors. The deposition temperature was kept at 380 °C. The
ALD process parameters for growing Ir are as follows: 6 s Ir(acac)₃ pulse/60 s N₂ purge/2 s O₂ pulse/6 s N₂ purge, resulting in a growth per cycle of 0.6 Å/cycle [17, 38]. For the present study, relaxation mechanisms such as scattering play an important role. Especially the scattering on grain boundaries and surfaces is influenced by the preparation method of the Ir films because the island formation depends on the growth conditions as detailed in [38].

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