Attosecond Photoscopy of Surface Plasmon Polaritons

Mattia Lupetti\textsuperscript{a}, J. Hengster\textsuperscript{b}, Th. Uphues\textsuperscript{b} and Armin Scrinzi\textsuperscript{b}

\textsuperscript{a}Physics Department, Ludwig Maximilians Universität, D-80333 Munich, Germany and
\textsuperscript{b}Center for Free-Electron Laser Science, Universität Hamburg, D-22761 Hamburg, Germany

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We propose an experimental arrangement which, with few modifications to state-of-the-art setups used for attosecond streaking experiments from solid surfaces, can characterize the transient temporal properties of Surface Plasmon Polaritons, in particular the excitation buildup time.

Surface Plasmon Polaritons (SPP) are collective excitations of electrons that propagate along a metal-dielectric interface. Recently, SPPs have been investigated for the development of ultra-fast all-optical circuitry \cite{1}, since they can combine the high operational speed of photonics (PHz scale) with the miniaturization provided by electronics (nm scale). For that purpose, it is important to understand the buildup dynamics and lifetime of the collective electronic excitation. Although the plasmon life-time can be inferred from the plasmonic resonance width (of the reflectivity spectrum, for instance), plasmon buildup is a process that cannot be addressed in terms of frequency analysis. From a theoretical point of view, having access to the transient dynamics of such excitations provides insight into the non-equilibrium physics of the collective motion.

We propose an experimental setup to measure the buildup time of a SPP, which can be realized with few modifications of the so-called “attosecond streak camera” setup \cite{2}, which has already been successfully applied to solid surfaces. The attosecond streak camera is a two-color pump-probe scheme, where a weak XUV attosecond pulse ionizes electrons from the solid, and a collinear, few-cycle (\(\sim 5\,\text{fs FWHM}\)) NIR pulse serves as the probe, which accelerates the XUV photo-electrons after their escape from the solid. With this technique, it was for the first time possible to resolve solid-state physics phenomena with resolution of few attoseconds (\(1\,\text{as} = 10^{-18}\,\text{s}\)) \cite{2}.

In our setup, the NIR pulse excites SPPs on a grating surface. A time-delayed XUV pulse probes the SPPs at different times during their evolution by detecting the effect of their evanescent fields on XUV photoemission. Different from atomic and surface streaking employed so far, the setup provides spatio-temporal information. Also, in principle, pump and probe beams can be spatially separated. To distinguish it from attosecond streaking experiments, we name our setup “Attosecond Photoscopy” of Surface Plasmon Polaritons.

A well established method for producing isolated attosecond pulses for ultrafast time resolved spectroscopy is the generation of high harmonic radiation (HHG) in noble gases \cite{3,4}. An intense few cycle NIR laser pulse is focused into a noble gas target and generates high harmonics of the fundamental radiation. The XUV radiation co-propagates with the driving laser pulse. Both pulses are focused onto a sample with a delayable two part mirror composed of a XUV multilayer mirror in the inner part and a broadband NIR mirror in the outer part. The multi-layer mirror is designed as a high pass filter for the harmonics, which results in an isolated attosecond pulse. The pulse can be timed relative to the NIR with a precision of \(\lesssim 10\,\text{as}\).

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Figure \(\text{1}\) illustrates the setup discussed here. The two beams propagate in \(y\)-direction, at normal incidence onto the plane of the grating. Polarizations are in \(x\)-direction, perpendicular to the grooves. Using this arrangement, two counter-propagating plasmons are excited in the focus of the NIR pulse on the grating structure. Depending on the time delay between the NIR pulse and the probing attosecond pulse, the XUV generated photoelectrons experience a different plasmonic field amplitude and phase leading to a modulation of the kinetic energy distribution by the emerging plasmonic field. Spatial information is gathered from the area covered by the XUV beam.

XUV photoelectrons are measured at perpendicular direction to the surface. As in \(\text{3}\), the final electron momenta are recorded as a function of the delay between the NIR and XUV beams. One obtains an electron spectrogram, which is a convolution of photoemission with acceleration in the plasmonic field at location and time of the initial electron release.

Below we analyze the photoscopic spectrogram using a basic analytical model as well as numerical solutions of

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1}
\caption{Experimental setup of an attosecond photoscopy experiment. The XUV attosecond pulse liberates electrons in the SPP fields, which is excited by a short NIR pulse. Control of NIR-XUV time delay enables the determination of SPP transient properties.}
\end{figure}
the SPP propagation together with a Monte Carlo simulation of the electron streaking process. We will demonstrate that one can recover from the spectrograms plasmonic buildup and life times. The spectrogram also images sub-NIR-cycle details of the NIR pulse, such as its carrier phase and, with proper adaptation of the measurement geometry, near-surface modulations.

Standard streaking experiments are based on electron sources that can be considered point-like with respect to the laser wavelength, such as atoms or molecules. For this reason the dipole approximation can be used: $A(r,t) \simeq A(t)$. After emission, the electron canonical momentum is conserved: $p(t) = p_i$, which translates into $\mathbf{p}(t) + \frac{e}{c} \mathbf{A}(t) = \mathbf{p}_i + \frac{e}{c} \mathbf{A}(t_i)$, where $e$ denotes the electron charge and $|\mathbf{p}_i| = \sqrt{2mE_{\text{XUV}} - W_f}$ is the initial momentum of the electron released at time $t_i$. Assuming that $A(t \to \infty) = 0$, the final momentum recorded by the spectrometer is

$$p_f = p_i + a(t_i),$$  \hspace{1cm} (1)

where we defined $a := \frac{e}{c} A$.

The spectral width of the XUV attosecond pulse is reflected in a momentum-broadening of the initial electron distribution $n_e = n_e(p_i,t_i)$. For simplicity we assume Gaussian distributions both, in time and momentum, which centered around a momentum $p_0$ and a time $t_0$, respectively. By $t_0$ we denote the time of peak XUV intensity on target. With Eq. (1) for the initial electron momentum, the time-integrated final momentum

$$\sigma(p_f) = \int_{-\infty}^{\infty} dt_i n_e(p_i,t_i) = \int_{-\infty}^{\infty} dt_i n_e(p_f - a(t_i),t_i).$$  \hspace{1cm} (2)

The spectrogram for a series of delays $\tau$ becomes

$$\sigma(p_f,\tau) = \int_{-\infty}^{\infty} dt_i n_e(p_f - a(t_i),t_i - \tau)$$  \hspace{1cm} (3)

From this, the NIR pulse can be reconstructed from an analysis of the average momentum of the streaking spectrogram:

$$\langle p(f) \rangle(\tau) = \int_{-\infty}^{\infty} dp_f p_f \sigma(p_f,\tau) \int_{-\infty}^{\infty} dp_f \sigma(p_f,\tau)$$  \hspace{1cm} (4)

If one detects only the electrons emitted parallel to the laser field, then the vector quantities in Eq. (4) become scalar. Taking $n_e(p_i,t_i) = N(p_0,dp) \times N(t_0,dt)$ as initial distribution function, where $N(a,b)$ is a normal distribution of mean value $a$ and variance $b$, one finds

$$\langle p(\tau) \rangle = -a(t_0 - \tau)$$  \hspace{1cm} (5)

which, up to a factor $e/c$, is the vector potential of the NIR streaking pulse. For this, we have considered XUV pulse durations $dt$ much smaller than the optical period of the NIR.

When applying the method to plasmonic excitations we have to consider that the SPP, acting as the streaking field, is spatially inhomogeneous and propagates on a surface, which is an extended electron source. Therefore, we need to include position dependence into our initial electron distribution: $n_e(p_i,t_i) \rightarrow n_e(r_i,p_i,t_i)$. The final momentum of the electrons accelerated in the plasmonic field is then

$$p_f = p_i - e \int_{-\infty}^{\infty} E(r(t'),t') dt'$$  \hspace{1cm} (6)

For a typical XUV photon energy of 80 eV, the average initial speed of a photoelectron is $v_i = 5 \text{ nm/fs}$. If the NIR source pulse is 4 fs short, it will give rise to a plasmonic field of a maximum duration of few tens of femtoseconds. During this time, the electrons move by $\lesssim 100 \text{ nm}$. The additional drift imparted by the plasmonic field is small compared to the initial velocity. As the plasmonic field extends to about NIR pulse wavelength (800 nm) beyond the surface, we can approximate it $r(t') \simeq r_i$ in Eq. (6). With this approximation, one obtains a position corrected analog of Eq. (1):

$$p_f = p_i - a(r_i,t_i)$$  \hspace{1cm} (7)

Since the photoelectron detector does not resolve the emission positions $r_i$, the photoscopic spectrogram is the integral over time and space

$$\sigma(p_f,\tau) = \int_{\mathbb{R}^3} d^3 r_i \int_{-\infty}^{\infty} dt_i n_e(r_i,p_f - a(r_i,t_i),t_i - \tau).$$  \hspace{1cm} (8)

For extracting time information from the photoscopic spectrogram, we use the delay-dependent momentum average and its variance

$$\langle p_f(\tau) \rangle = \frac{\int dp_f p_f \sigma(p_f,\tau)}{\int dp_f \sigma(p_f,\tau)}$$  \hspace{1cm} (9)

$$S(\tau) = \frac{\int dp_f |p_f|^2 \sigma(p_f,\tau)}{\int dp_f \sigma(p_f,\tau)} - \langle p_f \rangle^2$$  \hspace{1cm} (10)

As XUV pulse duration is short compared to the NIR period, we treat photoemission as instantaneous. The distribution of the photoelectron yield along the surface is proportional to the XUV intensity profile. Furthermore, we neglect any transport effect in the solid and consider only the photoelectrons coming from the first few layers of material as reported in [2]. With this one finds

$$n_e(r_i,p_i,t_i - \tau) \simeq g_s(x_i)n_e(p_i)\delta(y_i - y_g)\delta(t_i - \tau - t_0)$$  \hspace{1cm} (11)

where $y_g$ is the grating vertical position (we neglect any groove depth effect) and $g_s$ is a Gaussian function of width $w_g$, i.e. the XUV attosecond pulse focal spot.

As for the angular dependence of the photoemission we first restrict our discussion to the two extreme cases of 1)
unidirectional emission with all initial momenta orthogonal to the grating plane, or 2) isotropic emission. We will show later that the unidirectional case can be “filtered out” from the isotropic one by proper measurement arrangement. For either distribution, the reconstructed times closely reproduce the actual dynamics. In reality, the XUV photoelectron distribution will be between these extreme cases and should be determined in a measurement without NIR field.

Unidirectional initial distributions can be written as: $n_e(p_i) = n_e(p_i, \hat{n}_s)$, where $p_i = |p_i|$ and $\hat{n}_s$ is the direction orthogonal to the grating plane. Eq. (8) now for the initial electron momenta, we get:

$$\sigma(p_f, \tau) = \int_{-\infty}^{\infty} dx g_s(x_1) n_e (p_f - \hat{n}_s \cdot a(x_i, t_0 - \tau))$$

(12)

where $\hat{n}_s$ denotes the surface normal. Near the surface, in the region that is probed by the electrons, the SPP field is predominantly perpendicular to the surface. Therefore, we can approximate $\hat{n}_s \cdot a = a_y \approx a_{spp}$.

If we compute the momentum average and spread according to Eqs. (13), assuming a Gaussian distribution for the initial electron momenta, we get:

$$\langle p_f \rangle (\tau) = p_0 - \int_{-\infty}^{\infty} dx g_s(x_1) a_{spp} (x_i, t_0 - \tau) = p_0$$

$$S(\tau) = \Delta p^2 + \int_{-\infty}^{\infty} dx g_s(x_1) a_{spp}^2 (x_i, t_0 - \tau)$$

(14)

The spatial integral over the plasmonic vector potential vanishes, because the two counter-propagating have opposite polarizations, therefore $a_{spp}$ is an odd function of $x_1$. In general, few cycle pulses can be asymmetric, with the asymmetry depending on the Carrier-Envelop-Phase (φCEP). However, this asymmetry involves the complete spectrum of the NIR pulse, from which the plasmon resonance only cuts a comparatively small band. As a result, the observed sub-NIR beating of the signal reflects the absolute carrier phase, but is not sensitive to φCEP.

For isotropic XUV photo-electron emission, the initial distribution can be written as: $n_e(p_i) = \frac{1}{\pi} n_e(p_i)$, where we employed $p_i = |p_i|$ and $n_e = n_e(|p_f - a|)$. We use $|a| \ll |p_f|$ to approximate $|p_f - a| \approx p_f - a \cdot \hat{\theta}$, where $\theta$ is the angle between the final momentum and the surface normal. The spectrum then reads

$$\sigma(p_f, \tau) = \frac{1}{\pi} \int_{-\infty}^{\infty} dx g_s(x_1) n_e (p_f - a \cdot \hat{\theta})$$

(15)

A straightforward calculation for the angular integrations leads to the expressions for momentum average and variance

$$\langle p_f \rangle (\tau) = p_0 + \frac{1}{\pi p_0} \int_{-\infty}^{\infty} dx g_s(x_1) |a(x_i, \tau)|^2$$

$$S(\tau) = \Delta p^2 + \frac{1}{\pi} \int_{-\infty}^{\infty} dx g_s(x_1) |a(x_i, \tau)|^2.$$  

(16)  

(17)

Here $|a|^2 = a_x^2 + a_{spp}^2$ also includes $a_x$, the NIR field at the grating surface. Modifications of the surface NIR field compared to the incident beam can be measured in situ (see below).

In order to compute the spatial integrals in Eqs. (14-15), we consider a model plasmonic field of the form: $a_{spp} = \exp[i\varphi] \exp[-\varphi^2/\omega^2 T^2]$, with $\varphi = k_{spp} x - \omega t$. The total plasmonic field is assumed to be given by the two counter-propagating plasmons multiplied by a “buildup” and “decay” function of time only $a(t) = (1 - \cos(t/\xi)) \exp(-t/\Gamma)$, where $\xi$ and $\Gamma$ parameterize buildup and decay times, respectively. The plasmonic component of $a$ in $y$-direction is then

$$a_y(x, t) = a(t) \left[ e^{i\varphi_x} e^{-\frac{\varphi^2}{2 \omega^2 T^2}} - e^{i\varphi_y} e^{-\frac{\varphi^2}{2 \omega^2 T^2}} \right],$$

(18)

where $\varphi_{\pm} = \pm k_{spp} x - \omega t$ are the phases of the counter-propagating plasmons. Free parameters in this model are the SPP-pulse duration $T$, the excitation buildup time $\xi$ and the plasmon decay time $\Gamma$. The plasmonic term of the integrand in the momentum variance in Eqs. (14) is

$$S_{\tau} = a(\tau) \left[ 1 + e^{-\omega^2 T^2} - e^{-\frac{\varphi^2}{2 \omega^2 T^2}} \left( e^{-\omega^2 T^2} + \cos(2\omega \tau) \right) \right].$$

(19)

Simulations of the plasmonic field were performed with the finite-difference time-domain (FDTD) method, using a freely available software package [9]. As a pulse, we use a Gaussian 4 fs FWHM pulse at a central wave length of 800 nm. The grating was optimized for maximal absorption from the NIR pulse, assuming a gold surface. Beam waists of NIR and XUV were 5 and 10 µm, respectively. The XUV photoemission process is approximated as a sudden injection of electrons from the surface boundary, with the appropriate initial probability distribution function. The electron trajectories and final momenta are computed by solving the Lorentz equation for each photoelectron in the previously simulated electromagnetic field. The simulated spectrograms for both the unidirectional and isotropic energy initial electron distribution are shown in Figure 4.

From the photoclastic spectrogram we extract the momentum variance and fit expression (19) to the simulated photoclastic spectrograms. Results are reported in Table I. In all cases, the extracted buildup times are in good agreement with the times extracted directly from the FDTD simulations. The largest deviation is found for isotropic emission. However, that can be easily corrected by appropriate measurement geometry, where only electrons in the direction perpendicular to the surface are detected, with an opening angle of about 5 degrees (column named “filtered”). SPP pulse durations are most accurate for unidirectional emission, with acceptable errors for the other models. The recovery of lifetimes is poor, as the analytic model of the plasmon is correct only on short time scales up to about 30 fs. Considering
that lifetimes can be determined easily by spectroscopic methods, this is of secondary importance.

Selecting different detection angles gives access to further information. For example, measuring at a grazing angle in the polarization direction of the NIR gives direct access to the NIR field as in a standard streaking setup. A comparison of the two spectrograms in Figure 3 allows the evaluation of the field enhancement, which in this case is \( \sim 1 \). Moreover, performing a fitting procedure similar to what resulted in Eq. (19), we can also extract the time duration of the NIR pulse undergoing reflection from the spectrogram filtered at grazing incidence. Taking as model \( a(x, t) = \exp(-x^2/2w_0^2) \times \exp(-t^2/2\Delta t^2) \) and taking \( \theta = \pi/2 \) in Eq. (19), we get in lieu of Eq. (19) the following expression:

\[
S(\tau) = \Delta p^2 \left[ 1 + \frac{\Delta p^2}{(p_0 - \sqrt{2\pi} w_N \cos(\omega \tau)e^{-2x^2/\Delta x^2})^2} \right] (20)
\]

The fit to the simulated momentum variance gives a NIR pulse duration of \( \Delta t_{\text{FWHM}} = 4.5 \) fs, against the 4.6 fs measured in the FDTD code. Reconstruction of the NIR pulse undergoing reflection permits to evaluate the spectral distortion on the NIR pulse caused by the interaction with the grating surface.

In conclusion, we have shown how one can image, with existing experimental means, buildup, duration, and lifetime SPP excitations. We found the reconstruction of these parameters to be robust against various scenarios of photo emission and experimental arrangement. The same setup also provides \emph{in situ} diagnostics of the NIR pulse. Once spatially separated XUV attosecond and NIR pulses become available, one may resolve in space and time also other surface phenomena: by letting the NIR field excite a surface mode in some region, one can image SPP propagation along complex plasmonic waveguides or plasmonic switches by simply pointing the attosecond XUV pulse on the region of interest.

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**TABLE 1.** SPP-pulse duration \( T \) and buildup time \( \xi \) resulting from fits of the theoretical model on the numerically simulated data. The cases isotropic emission, isotropic emission with unidirectional detection (“filtered”), and unidirectional emission, as well as values extracted directly from the FDTD calculation are shown. (Times in fs.)

|          | Isotropic | Filtered | Unidirectional | FDTD   |
|----------|-----------|----------|----------------|--------|
| \( T \)  | 29.0 \pm 2.5 | 20.7 \pm 0.6 | 25.50 \pm 0.75 | 25.2 \pm 0.1 |
| \( \xi \) | 3.3 \pm 0.2 | 3.61 \pm 0.06 | 3.68 \pm 0.05 | 3.6 \pm 0.1 |
| \( \Gamma \) | 48 \pm 5 | 77 \pm 4 | 60 \pm 3 | 90.0 \pm 1.0 |

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**FIG. 2.** Comparison of the photoscopic spectrogram for an initial broad energy unidirectional (left) and isotropic (right) electron distribution. In the isotropic case, the buildup signature is modulated by transverse streaking at \( \omega \) frequency (cf. Eq. (20)). The overlapping SPPs beat at \( 2\omega \) (cf. Eq. (19)), whose phase is locked to the carrier phase of the incident pulse.

**FIG. 3.** Comparison of the photoscopic spectrograms obtained by measuring the photoelectron final momenta with a TOF detector of 5 degrees acceptance angle placed respectively at orthogonal direction (left) and in polarization direction (right). The measurements retrieve plasmonic, and NIR field, respectively.
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