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High-temporal-resolution electron microscopy for imaging ultrafast electron dynamics

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I. The electron-photon coupling between the NIR laser pulse and the original few-hundred femtosecond electron pulse

For retrieving the cross-correlation temporal profile of the gated electrons coupling with the NIR pulse (Fig. 3E, in main text) from the cross-correlation temporal profile of the NIR laser pulse coupling with both original electron and gated electron pulses, the measurement of the NIR pulse coupling only with the original electron pulse is required. Therefore, the electron energy spectrum as a function of the NIR laser pulse delay (τ_{NIR}) (no gating pulse) is recorded and the average of 5 scans is plotted in Fig. S1a (the ZLP has been subtracted from the spectrogram for better display of the coupling peak). The total electron energy spectrum at $\tau_{NIR} = 0\, fs$ is shown in Fig. S1b. The cross-correlation temporal profile is extracted by calculating the integration of the coupling peaks in each spectrum at τ_{NIR} and plotted in Fig. S1c. The cross-correlation temporal profile of the gated electron pulse can be obtained by subtracting this extracted profile and the temporal profile of the cross correlation between the NIR pulse and both original and gated electron pulses (Fig. 3e, in main text). The coupling efficiency between the NIR and the original electron pulse is on the order of ~11%, which is above the saturation ~7%, thereby causing a broadening in the measured cross-correlation temporal profile between the gated electron and NIR laser pulse (Fig. 3e, in main text).

II. Attosecond optical gating of electron pulse

To explore the practicality of attosecond optical gating, we have conducted a simple theoretical calculation of the efficiency of electron coupling with the optical attosecond pulse demonstrated in ref (39). This has been done by calculating the probability of one electron emitting or absorbing photons through interaction with the surface of a nanostructure following the steps outlined in ref (39). Briefly, the electron-photon interaction is mediated by an evanescent electromagnetic field induced by the optical attosecond gating photon pulse hitting the surface. The spatial distribution of the evanescent field is determined by the optical property and geometry of the nanostructure. For simplicity, and without losing the physical essence, we assume an effective one-dimensional spatial distribution of the evanescent field.

Then, the strength of the evanescent field coupled with the electrons is given by

$$E_z(z,t) = E_0 e^{-|z|/\xi} e^{-i\omega t-(t+\tau)^2/\tau_p^2} \qquad (1)$$

where E_0 is the peak strength of the evanescent field, ξ is the penetration depth into the vacuum, ω is the photon frequency, τ is the delay between the optical and electron pulses, and τ_p is the duration of the optical pulse. The temporal evolution of the electrons in the presence of the evanescent field can be described by a time-dependent Schrödinger equation including a light-matter coupling Hamiltonian with a classical electromagnetic field, whose solution can be found self-consistently by solving the Lippmann-Schwinger equation (Eq. (4) in ref (39)). Expanding the electron wave function in a basis of momentum eigenstates corresponding to gaining or losing a certain number of photons, the Lippmann-Schwinger equation can be solved, and the expansion coefficients are given in a recursive manner (Eq. (9) in ref (39)). With the expansion coefficients, the probabilities of electrons gaining/losing L photons can be computed from the equation (1) in the main text.

Using this model, we have calculated the coupling probability of the optical attosecond gating pulse, which is already demonstrated[11] and a ~75 fs electron pulse which can be obtained by conventional electron pulse compressor techniques.[17-19] The evanescent field intensity on the surface of the thin film is assumed to be 1.5×10^{12} W/m^2, with a penetration depth of 200 nm into the vacuum. Then, the calculated probability has been used to calculate the coupling spectrogram, which consists of the calculated coupling energy spectrum as a function of the delay (τ_{OAP}) between the electron and optical gating pulse (Fig. 4b, in the main text). The coupling peak is broader due to the spectral broadening of the gating pulse.

The attosecond optical gating efficiency can be obtained by dividing the number of gated electrons (N_g) by the total number of electrons (N_t). The relation between the evanescent field intensity and the efficiency of the attosecond optical gating is calculated and plotted in Fig S2. From this intensity dependence calculation, we have chosen the evanescent field intensity at low value (1.5×10^{12} W/m^2) in the liner regime (Fig S2) to avoid the broadening effect due to saturation in the optical attosecond gating probability and efficiency calculation in Fig. 4 in the main text.

The gating efficiency of 75 fs electron pulse by the optical attosecond pulse is on the order of ~1%. This efficiency increases for the gating of shorter original electron pulse. The attosecond optical gating efficiency for different original electron pulse durations is calculated and plotted in Fig. 4D in the main text. This calculation shows that the number of electrons that can be gated in an attosecond time window can achieve 8% of the total number of electrons in an original electron pulse with a duration of 10 fs. This demonstrates the ability of the optical gating approach to be used for generating subfemtosecond electron pulses with sufficient strength to image the electronic dynamics.

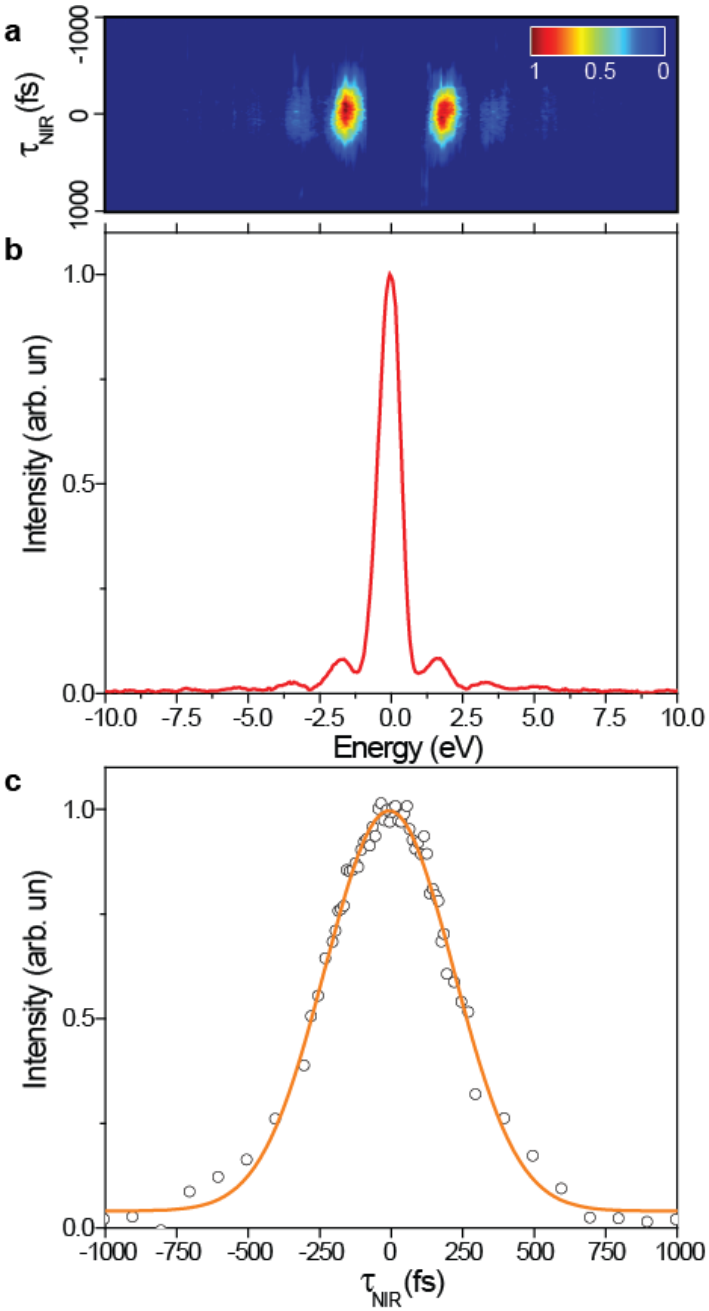

Fig. S1. Electron-photon coupling between NIR and original electron pulses. **a**, The measured electron energy spectrogram as a function of the NIR pulse delay (τ_{NIR}). The ZLP has been subtracted from the spectrogram. **b**, The full electron energy spectrum at $\tau_{NIR} = 0\ fs$. **c**, The retrieved cross-correlation temporal profile calculated from the measured spectrogram in (**a**).

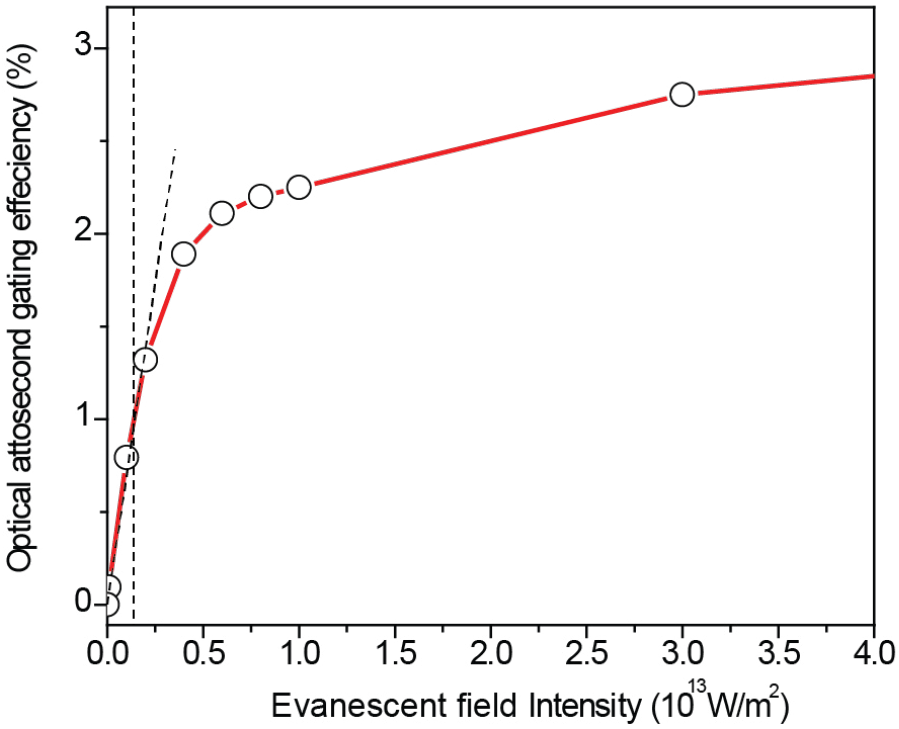

Fig. S2. The relation between the evanescent field intensity and the optical attosecond efficiency.