Supporting Information for:

All-optically Reconfigurable Plasmonic Metagrating for Ultrafast Diffraction Management

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1. Design of the dimeric metaatom

The hybridization theory design of the unit cell of the plasmonic metagrating was supported by full-wave numerical analysis with a commercial tool (COMSOL Multiphysics 5.4). A 2D model implementing Maxwell’s equations in the frequency domain according to the scattered field formalism has been developed to characterize the optical behaviour of the individual nanostrips and of the nanostrip dimer surrounded by a homogeneous non-dispersive and lossless medium with effective permittivity $\varepsilon_{\text{eff}}$. For the static permittivity of gold, $\varepsilon_{\text{Au}}(\lambda)$, we considered the analytical model from Ref. 3 fitted on the experimental data from Johnson and Christy.4 A spectral sweep analysis was conducted to determine the scattered electric and magnetic vector fields and evaluate absorption, scattering and extinction cross-sections of the metallic dimer. Perfectly matched layers (PMLs) with scattering boundary conditions (BCs) were set to avoid numerical artifacts at the boundaries of the circular domain embedding the structure. Far-field COMSOL feature, implementing Stratton-Chu formulas5 on a surface enclosing the free-standing dimer, has been employed to evaluate the dimer far-field scattering angular pattern.

2. Hybridization scheme validation

To discuss the behaviour of the dimeric metaatom in its disconnected configuration, a hybridization scheme based on the response of the single nanostrip monomers (refer to Fig. 1c from the main text) has been employed. To validate this view, numerical simulations have been performed according to above section 1 in order to determine the spatial distribution of the induced charge density. The main results of the computation at the two resonance wavelengths are presented in Fig. S1, showing the norm of the scattered electric field $E_S$ (normalised to the incident field, $E_0$) across the structure. Based of the electric field spatial patterns, charge density can be straightforwardly deduced by invoking the Gauss’ law: regions where the divergence of $E_S$ is higher correspond to higher induced charge density.
Appropriately, circles in Fig. S1 highlight such regions. In particular, red circles surround portions of the dimers with positive charge density, whereas white circles indicate regions with negative charges. Note that the distribution of induced charge density in the dimer retrieved by full-wave numerical analysis well compares, qualitatively, with the scheme of charges reported in Fig. 1c, apart from unavoidable quantitative discrepancy due to retardation effects and flip of sign (having no relevance). Hence, the numerical investigation endorses the hybridization scheme of the dimeric metaatom and ascertains the origin of the two resonant peaks observed in the optical response of the dimeric structure.

Figure S1: Spatial pattern of the dimeric metaatom optical excitation. a-b, Spatial map of the scattered electric field enhancement $E_S/E_0$ (in modulus) across the 2D dimeric nanostrips metaatom, evaluated at the excitation wavelengths $\lambda = 930$ nm (a) and $\lambda = 575$ nm (b). Black arrows indicate the scattered field lines of force. Circles highlight the regions where electric field divergence is higher (red for positive, white for negative sign), corresponding to a higher induced charge density.

3. Ultrafast photoexcitation modelling

As concisely outlined in the main text, the modelling of the photoinduced inhomogeneous distribution of hot carriers, which results in the symmetry breaking enabling the diffraction management in plasmonic metagratings, consists of several aspects. To simulate the interaction of the plasmonic metagrating with both the control pulse (according to the I3TM), and
the signal pulse, a suitable finite element method (FEM)-based 2D model has been developed employing COMSOL Multiphysics, and a segregated approach was pursued.

3.1 Pump absorption pattern calculation

First, the scattering problem in the frequency domain is solved in static conditions to determine the absorption spatial pattern of the control pulse. A monochromatic \( p \)-polarized plane wave at \( \lambda_c = 600 \text{ nm} \) impinging at \( 45^\circ \) is assumed (Fig. 3a). Unperturbed Au permittivity \( \varepsilon_{\text{Au}}^0(\lambda) \) has been considered for the plasmonic metaatoms. Periodic BCs have been set to simulate the array optical behaviour and ports formalism is employed to compute the near fields and far fields of the structure. This step provides us with the key quantity \( A(\vec{r}) \) to evaluate the space dependence of the absorbed power density (Eq. (1) in the main text) and to formulate the I3TM from its source term.

Starting from the latter result, the drive term of the ultrashort pulse illumination dynamical model, \( P_{\text{abs}}(\vec{r}, t) \), can be addressed. Note that such a formulation for the absorbed power holds in a linear regime, where the control pulse fluence is such as not to self-induce nonlinear interactions. This may lead to a quantitative underestimation of the absorbed power, although with no essential modifications in the photoexcitation dynamics for the relatively low fluence here considered.

3.2 Solving the I3TM

The core of the energy transfer dynamics is then the I3TM of Eqs. (2)-(4) from the main text, that we developed and used to describe the hot-electron driven symmetry breaking and predict the diffraction management effect. The model is a generalization of the well-established and widely employed 3TM, where mainly two aspects have been introduced.

First of all, the source term \( P_{\text{abs}}(\vec{r}, t) \) has been considered as a space-dependent quantity, hence inducing an inhomogeneous distribution, via Eq. (2), of the energy density \( N \), which relates to the variation of the occupation probability of electrons. Further, spatial Fourier-
like diffusion terms for both the electronic and the lattice temperatures have been included in the model, Eqs. (3)-(4), so to describe the propagation in space of the thermal non-uniform perturbation in electron and phonon populations in the plasmonic nanostructures.

All the coefficients of the rate coupled partial differential equations of the I3TM are deduced, straightforwardly or by direct comparison, from more formal and fundamental models of ultrafast photoexcitation of metallic nanostructures (see e.g. Refs. [8–12 for further details, at the basis of the formal foundation of our I3TM, not discussed in the present work for the sake of conciseness).

In particular, regarding the parameters in the model Eq. (2), \( a \) and \( b \) coefficients govern the time decay of the energy density stored in the nonthermalised fraction of the electronic gas, \( N(\vec{r},t) \), following electron-electron \((e-e)\) and electron-phonon \((e-ph)\) scattering events, respectively. By referring to the corresponding scattering times \( \tau_{e-e} \) and \( \tau_{e-ph} \), out-of-equilibrium electrons heating rate is expressed as the average value \( a = \langle 1/\tau_{e-e} \rangle = \hbar \omega_c / 2\tau_0 E_F^2 \), where \( \hbar \omega_c \) is the control pulse photon energy, the empirical time \( \tau_0 \) has been set equal to 13.5 fs\(^{13,14}\) and \( E_F \) stands for the Au Fermi energy. The energy transfer from the nonthermal electron gas to Au phononic temperature occurs at rate \( b = 1/\tau_{e-ph} = k_B \Theta_D / t_f \hbar \omega_c \), with \( k_B \) the Boltzmann constant, \( \Theta_D \) the Au Debye temperature, \( t_f \) the quasi-particle free flight time.\(^{8,9}\)

When dealing with Eqs. (3)-(4), \( C_e \) and \( C_l \) refer to the specific heats of the thermal electrons and lattice phonons respectively, and \( \kappa_e \) and \( \kappa_l \) are their thermal conductivities, while \( G \) stands for the thermal electron-phonon coupling term. The thermal properties of the lattice have been considered as constant;\(^{15,16}\) \( C_l = 2.49 \cdot 10^6 \) J m\(^{-3}\) K\(^{-1}\) and \( \kappa_l = 317 \) W m\(^{-1}\) K\(^{-1}\). On the other hand, due to the significant increase of \( \Theta_e \), electronic thermal properties as well as the coupling parameter \( G \) have been modelled as electronic temperature-dependent: \( C_e(\Theta_e) \) and \( G(\Theta_e) \) are computed starting from the total density of electronic states, in agreement with Ref. \(^{12}\); electronic conductivity is expressed as\(^{15,16}\) \( \kappa_e = \kappa_l \Theta_e / \Theta_l \).

In terms of the model numerical implementation, with a subsequent time-domain analysis
we solved Eqs. (2)-(4) to determine the local temporal dynamics of the three energetic degrees of freedom in each point of the plasmonic structure. Both time (by fixing the time step \( dt = \Delta t/30 \)) and space (by setting the mesh maximum element size to \( r/2 = 5 \) nm) have been appropriately discretized to resolve the photoexcitation spatio-temporal evolution. This step retrieves the spatial distribution of \( N, \Theta_e \) and \( \Theta_l \) in time, which is then used to compute the corresponding Au permittivity modulation pattern, \( \Delta \varepsilon(\vec{r}, \lambda, t) \), that presides over the optical symmetry breaking of the plasmonic metaatoms.

### 3.3 Calculation of photoinduced permittivity changes

Regarding the calculation of the photogenerated inhomogeneous permittivity modulation, as mentioned in the main text, contributions from \( N, \Theta_e \) and \( \Theta_l \) have been considered in describing both the intra- and interband terms of Au permittivity, based on well-established models of the metal nonlinear optical properties upon ultrashort pulse illumination.

For the intraband Drude-like contribution, this term has been considered to be modified by a change in \( \Theta_e \) and \( \Theta_l \) via the Drude damping \( \Gamma \) and the metal plasma angular frequency \( \omega_p \). In particular, \( \Gamma \) has been written as the sum of an electron-electron \( \Gamma_{e-e} \) and an electron-phonon \( \Gamma_{e-ph} \) damping factor. The former is derived in the framework of Landau’s Fermi liquid theory and exhibits a quadratic dependence on \( \Theta_e \), the latter can be evaluated from the Fermi golden rule at the second order and, due to the limited ranges of \( \Delta \Theta_e \) predicted, approximated to be directly proportional to \( \Delta \Theta_l \) and quadratically to the control pulse photon energy. The plasma pulsation modification instead has been considered to be driven by the volume expansion of the metal nanostructure. Its modification has been formulated in accord with Refs. [21, 23].

On the other hand, the interband modulation is driven by both nonthermal and thermalised electrons, and has been determined based on the description of the nonlinear thermomodulation in Au originally proposed by Rosei and co-workers, who analytically linked the changes in energy distribution of carriers to the optical transition rate between valence...
and conduction bands. By following the formulation reported in Refs. [9,27] by one of the present authors, the effect driving such modification in absorption, directly affecting the permittivity imaginary part $\varepsilon''$, is the so-called Fermi smearing, which occurs when conduction electrons are brought out of equilibrium following photoexcitation and resulting heating of the electronic distribution. A proper description of such effect leads thus to an analytical expression for $\Delta \varepsilon''$, its corresponding real part, $\Delta \varepsilon'$, being readily retrieved via Kramers-Kronig analysis.

Figure S2: Spectrally-dispersed permittivity modulation in space and time. Results of the numerical calculations of the nonlinear photoinduced Au permittivity modulation spectra in different points of the considered metaatoms at different control-signal pulses delays. **a-c**, From left to right: spectra of the real part of the permittivity change, $\Delta \varepsilon'(\vec{r}, \lambda; \tau)$, at $\tau = 100$ fs, $\tau = 2$ ps and $\tau = 20$ ps, respectively, evaluated at two exemplary points of the Au metaatom, for both the disconnected (dashed lines) and connected (solid lines) configurations, as shown in the right inset. **d-f**, Same as (a)-(c) for the imaginary part of the permittivity change, $\Delta \varepsilon''(\vec{r}, \lambda; \tau)$.

Figure S2 shows in detail the spectra of both the real (upper panels) and imaginary (lower panels) parts of the spectrally-dispersed permittivity change in two exemplary points of the plasmonic structure (highlighted in the inset). Such modification, defined locally and
evolving in time, is evaluated at different control-signal pulse delays, revealing the origin of the optical symmetry-breaking effect driving the ultrafast diffraction management we here report. Indeed, at short delays, each point is optically characterised by a different value of $\Delta \varepsilon$, which makes the unit cell of the metagrating non-symmetric as in static conditions and breaks the degeneracy of the $\pm 1$ diffraction orders. This happens in both the considered dimer configurations, namely when the two nanostrips are disconnected (dashed lines) as well as connected (solid lines). However, for longer time delays, the two systems exhibit a significantly different behaviour, explaining the dramatic differences in terms of diffraction management already highlighted in Figs. 4 and 5 of the main text. Concerning the disconnected structure, spectra of $\Delta \varepsilon$ evolve in time according to the temporal dynamics of the energetic degrees of freedom driving the optical perturbation (refer to Section 6 for further details). Nevertheless, the curves evaluated in the considered points remain separated even at $\tau = 20$ ps, keeping the metaatom asymmetric from the optical perspective.

On the other hand, dynamics of the spectra change considerably for the connected configuration. By referring to the solid curves in panels (c) and (f) of Fig. S2, curves are folded, as a result of the collapse of the local inhomogeneities. Following hot electron diffusion and electronic temperature homogenisation (much more relevant when compared to effects related to lattice temperature), the symmetry is restored and the permittivity is again single-valued within the whole plasmonic metaatom. This closes definitely (or up to a second control pulse possibly impinging on the metagrating even in the THz regime) the degeneracy breaking of $\pm 1$ diffraction orders (refer to the trend of $D_R$ and $D_T$ in Fig. 5 and Fig. S5). However, note that, even in the connected configuration, metaatoms are still strongly excited at longer time delays and the signal pulse would experience a non-zero $\Delta \varepsilon$ up to tens of ps. The two curves of permittivity change in the two nanostrips overlap, but are much different from 0, which represents the static condition. This facet encloses the core of the ultrafast nature of the diffraction management discussed in the present work, which overcomes the speed bottlenecks of hot-carrier-based nanophotonics applications imposed by electronic relaxation
times.

Regarding the evolution of permittivity spectra, the predicted changes in sign, values and the main dispersion features here shown are consistent with calculations reported and validated in other previous works dealing with the homogeneous 3TM (see e.g. Refs. 9,28). Such features belong to the non-trivial interplay of the broadband contributions arising from nonthermal and thermal hot electrons, which govern the overall optical permittivity modulation of gold nanostructures excited with ultrashort laser pulses. Note that the zero crossing of real part permittivity change is achieved at around 550-570 nm during the initial steps of the dynamics (Figs. S2a and S2b), and then blue shifts to 510 nm for longer delay of 20 ps (Fig. S2c). This behaviour is in good agreement with the zero-crossing observed in the transient $D_R$ spectra of Fig. 3d for increasing time delay.

### 3.4 Modulation of metagrating diffraction

To finally determine the effect on the metagrating optical diffraction orders of the photoinduced inhomogeneous perturbation experienced by a signal pulse, a frequency-domain simulation is then performed over a broad spectral range for a given control-signal delay $\tau$, which acts as a parameter. Numerical implementation is similar to the first step (Section 3.1), however Au permittivity is modified according to the previous step (3.3) and given by $\varepsilon_{Au}(\vec{r}, \lambda; \tau) = \varepsilon_{0}^{\text{Au}}(\lambda) + \Delta \varepsilon(\vec{r}, \lambda; \tau)$ and the $p$-polarized plane wave modelling the signal pulse impinges on the structure at normal incidence. All the array diffraction orders are then computed via port formalism at different time delays.

Note that the linear and segregated approach we pursued in modelling control-signal pulses interaction with the plasmonic metagrating is licit as long as the control pulse has a time duration such that several optical cycles are contained in the pulse (that is the so-called Slowly Varying Envelope Approximation$^{29}$). In light of the parameters considered in the simulations, the hypothesis is well fulfilled, making the model reliable in the considered excitation conditions.
To quantify the transient perturbation photoinduced by the control pulse, spectra of the diffraction orders, computed for the excited metagrating at a given time delay, are then compared with the corresponding unperturbed values. This allows one to determine the dynamical evolution of the spectra for all the diffraction orders supported by the periodic structure. In the case under investigation (both in the disconnected and the connected configurations), these are the direct transmission ($T_0$) and reflection ($R_0$) orders, the ±1 orders both in transmission ($T_{±1}$) and reflection ($R_{±1}$) and the $T_{±2}$ orders (albeit much weaker), along with absorption $A$ (cf. Figs. 2 and S4).

Figure S3: **Transient modulation of the metagrating optical response.** Spectra of the all-optical modulation of each of the diffraction orders supported by the periodic structure, here shown for the disconnected metagrating configuration at a fixed control-signal time delay of $\tau = 100$ fs. Variations are shown for the ±1 orders both in transmission and reflection (a), and for direct transmission $T_0$ and reflection $R_0$, along with absorption $A$ (b). c, Spectra of the sum of the modulations either in the ±1 orders shown in (a) or in $T_0$, $R_0$ and $A$ from (b) are compared to show power redistribution and conservation.
Although the present work has been mostly focused on the modulation of ±1 diffraction orders, since those are the ones responsible for a symmetry breaking in the metagrating optical response, it is worth highlighting that the control pulse induces in fact an all-optical modulation for each of the aforementioned orders. Throughout photoexcitation, a variation evolving in time occurs for each of them, since ultrashort radiation entails a modification of Au optical properties, regardless of the symmetry arguments which hold for the ±1 orders. Interestingly, the fact that the control pulse can indeed change dynamically the spectra of $T_0$, $R_0$ and $A$ introduces some extra channels in the framework of the interaction between the signal pulse and the photoexcited metagrating at a certain delay $\tau$. In other terms, the power of the signal pulse can be redistributed not only between reflection and transmission involving exclusively the ±1 diffraction orders. Therefore, for a given $\varepsilon_{Au}(\vec{r}, \lambda; \tau) = \varepsilon^0_{Au}(\lambda) + \Delta\varepsilon(\vec{r}, \lambda; \tau)$, absorption as well as transmission and reflection 0-th orders are also modified, upon the constraint of total power conservation, that is the optical theorem, which requires that the sum of all orders and losses equals one, corresponding to all order variations summing to zero.

This is clearly shown in Fig. S3, where modulation spectra of all the orders involved in the structure optical response are reported in the case of the disconnected configuration at an exemplary time delay of $\tau = 100$ fs. While Fig. S3a shows the spectra of the ±1 diffraction orders, more extensively discussed in the work, the spectrally-dispersed variations for absorption losses and direct transmission and reflection are reported in Fig. S3b. According to power conservation (or, equivalently, the optical theorem written for periodic structures), all these quantities sum to zero, as confirmed in Fig. S3c, where perfectly specular spectra, computed as the sum of curves in Figs. S3a and S3b respectively, are depicted (purple and green curves, respectively). Note that there is a slight mismatch, at short wavelengths, in the zero-crossing of curves in Fig. S3c. This difference is due to the contribution of the $T_{\pm 2}$ orders, accounted for in numerical simulations, but not included in the sum here presented for the sake of clarity. This couple of orders is in fact non-zero only in the blue region of
the spectrum, with an output power which is more than one order of magnitude weaker if compared to any other.

Generally speaking, full-wave simulations of the metagrating optical response reveal a complex dynamics of diffraction from the periodic structure. Spectrally-dispersed variations of all the diffraction orders are photoinduced by the ultrashort control pulse, which modifies the optical properties of the metaatom. As a result, throughout the interaction between the signal pulse and the illuminated metagrating, the all-optical diffraction modulation involves all the available channels for power redistribution. In particular, the $R_{\pm 1}$ and $T_{\pm 1}$ orders compete with modulation of direct transmission and reflection as well as absorption losses, experiencing a non-zero modulation as well, although with no role in the symmetry-breaking mechanism reported in this work.

4. Optical static response of the connected configuration

The plasmonic 1D array with metaatoms made of pairs of connected nanostrips was first analysed by calculating the static optical response of the resulting metagrating. The same numerical tools presented in above sections and employed to discuss the disconnected metaatom configuration (refer to Fig. 2) have been used by consistently changing the modelled geometry.

![Figure S4: Plasmonic metagrating with connected metaatom configuration. a, Unit cell defined for 2D simulations. b-c, Static optical response of the array in terms of (b) absorption and zero order of reflection and transmission and (c) first (+1 and −1) diffraction orders upon linearly $p$-polarized optical excitation at normal incidence.](image)
While, as discussed in the main text when commenting on Figs. 4 and 5, such a slight topological modification has a dramatic impact on the temporal evolution of the quantities of interest, it only marginally affects the static optical behaviour of the structure. In particular, by comparing Figs. S4b-S4c and Figs. 2b-2c, the computed spectra do not reveal any substantial qualitative difference. When excited by a linearly $p$-polarized (electric field along the $x$-axis) plane wave at normal incidence, the connected nanoantenna array exhibits a well-defined resonant peak in the zero order transmission (blue curve in Fig. S4b) close to 590 nm, exactly as in the case of disjointed nanostrips. Furthermore, similarly to its disconnected counterpart, $+1$ and $-1$ orders, both in transmission and reflection, are non-zero, as depicted in Fig. S4c. For the same symmetry reasons as for the disconnected metaatoms, such $\pm 1$ orders are degenerate as well, enabling the all-optical ultrafast diffraction management reported in this work.

Note that, in modifying the topology of the single metaatom, all material properties and geometrical parameters have been kept constant but $W$, i.e. the longitudinal dimension of the individual nanostrip. Indeed, $W$ has been set equal to 150 nm (instead of its original value, 165 nm) to keep the single dimer metallic volume constant between the two configurations (see Fig. S4a). In such a way, we ensured a fair comparison between the two topological variants of the metaatom.

5. Diffraction management in the connected configuration

As for the disjointed dimeric nanoantenna array detailed in the main text, all-optical diffraction management of the connected variant has been predicted and investigated by exciting the array with a control pulse impinging at $45^\circ$ on the structure and by probing the resulting optical perturbation with a signal pulse, arriving at normal incidence at delay $\tau$.

The ultrafast symmetry breaking effect, as for the disconnected configuration, is then evaluated by considering the evolution in time of the spectra for the $\pm 1$ reflection (transmission)
orders, as well as the figure of merit $D_R$ ($D_T$) introduced in the main text, Eq. (5), and measuring the optically-induced asymmetry between originally degenerate diffraction orders.

In particular, Figure S5 compares spectra of reflection ($R_{\pm 1}(\lambda, \tau)$ in Figs. S5a, S5c) and transmission ($T_{\pm 1}(\lambda, \tau)$ in Figs. S5b, S5d) $\pm 1$ orders at different delays (dashed lines for $\tau = 100$ fs, dotted lines for $\tau = 2$ ps) with their respective static (degenerate) cases (solid lines, $\tau = 0$). Such a direct comparison further stresses the effect of the topological change in the nanoantenna. Indeed, spectra at $\tau = 100$ fs can be interpreted in the same terms as the results for the disjointed configuration (refer to Figs. 3b, 3c). The inhomogeneous absorption pattern of the control pulse (moreover very similar in the two considered variants) induces, via $N, \Theta_e$ and $\Theta_l$, a non-uniform optical perturbation $\Delta \varepsilon(\vec{r}, \lambda; \tau)$ across the structure, resulting in a symmetry breaking of the $\pm 1$ orders following the ultrafast photoexcitation.

On the other hand, the dynamical trend of diffraction orders is dramatically different in the two configurations after few ps. While spectra at $\tau = 2$ ps for the disconnected structure

![Figure S5: Ultrafast diffraction management. a, First order transient reflection spectra $R_{\pm 1}(\tau)$ at $\tau = 100$ fs time delay (dashed lines) after control pulse absorption, compared with static (and degenerate) reflection spectra $R_{\pm 1}^0$ (solid lines). Optical excitation conditions (polarisation and incidence angle of both the control and the signal pulses) are the same as in Fig. 3 in the main text. b, Same as (a) for transmission orders. c-d, Same as (a)-(b) for a time delay of $\tau = 2$ ps. e, Spectra of the $D_R$ figure of merit for optical symmetry breaking in reflection, at different delays $\tau$. f, Same as (e) for transmission $D_T$ figure of merit.](image-url)
are essentially equivalent to the ones reported in the main text, Figures S5c-S5d show the optical effect of electronic homogenisation across the structure. Evidently, the system is still strongly out of equilibrium, since $R_{\pm1}(\lambda, \tau)$ and $T_{\pm1}(\lambda, \tau)$ (dotted lines) are much different from their corresponding static spectra (solid lines). However, they do overlap again, as a consequence of the optical symmetry recovery, occurring in about 2 ps, well before relaxation.

Analogous considerations can be drawn by analysing the time evolution of the symmetry-breaking figures of merit in Figs. S5e-S5f. The overall trend at short control-signal pulses delay is similar to the separated nanostrips configuration (refer to Figs. 3d-3e). Nevertheless, in the connected topology both $D_R$ and $D_T$ are almost completely back to zero at $\tau = 2$ ps, thanks to the electronic diffusion across the whole metaatom, enabling ultrafast symmetry recovery and subsequent restoring of $+1$ and $-1$ diffraction orders degeneracy.

6. Dynamics of the internal degrees of freedom

In comparing the two, disconnected and connected, configurations of the metagrating under investigation, it is apparent that the key feature governing the optical symmetry breaking is thermal equilibration between the two strips in the metaatom. In particular, the recovery of the power balance between $\pm1$ diffraction orders is accomplished when electron temperature $\Theta_e$ and, although to a lesser degree, Au lattice temperature $\Theta_l$, recover a symmetric spatial distribution. Equilibration of electronic temperature in the two strips of the metaatom occurs via two channels, namely electron-phonon relaxation processes and hot-carrier diffusion. Although both present in the disconnected configuration as well, the gap between strips inhibits the energy flow, whereas the behaviour of the electrically connected metaatoms is indeed mostly dominated by the latter.

Such a fundamental difference in the dynamics of the optical response from the two configurations can be expressed and seized in terms of the time evolution of the internal energetic degrees of freedom for the plasmonic metaatom, as reported in Fig. S6. These are the excess
Figure S6: **Dynamics of internal energetic degrees of freedom.** Temporal dynamics of the excess energy density in nonthermal electronic population (a), the hot carriers temperature variation (b) and the lattice temperature variation (c), shown in two exemplary points (refer to the panel insets) of both the disconnected (dashed lines) and connected (solid lines) metaatoms.

of energy stored per unit volume in the nonthermal fraction of the electronic population, $N$ in Fig. S6a, the temperature of electrons in the metal Fermi sea, $\Theta_e$ in Fig. S6b, and the lattice temperature $\Theta_l$ (in Fig. S6c). Temporal dynamics in two representative points (refer to Fig. S6 right inset) across either the disconnected (dashed lines) or the connected (solid lines) metaatom geometry are compared over a long (up to 20 ps) time delay range. $N(\vec{r}, t)$ follows the typical temporal dynamics solution of the rate equations written for ultrafast light-matter interaction (Eqs. (2)-(4) in the main text), decaying due to scattering events with a characteristic time of $\sim 350$ fs, in agreement with more accurate theoretical results.\(^{7,9}\)

Moreover, since according to the I3TM their distribution in space inherits the spatial pattern of the control pulse absorption\(^{30}\) ($A(\vec{r})$ in Eq. (1)), which is similar for the two configurations at $\lambda_c$, the dynamics of $N$ across the disconnected and the connected metaatoms are comparable. Therefore, in terms of contribution to the optical perturbation arising from nonthermal carriers (refer to Section 3.3) and corresponding transient symmetry breaking, the structures are expected to behave alike. On the other hand, the considered metaatom geometries exhibit deeply different behaviours when both electronic and lattice temperatures are considered. In fact, $\Theta_e$ follows qualitatively comparable dynamics in the four considered points, showing that a full relaxation of the electronic excitation requires some tens of ps. The major difference between connected and disconnected structures is, as highlighted in commenting
on Fig. 4c in the main text, given by homogenisation, achieved in the connected structure only. Besides and, most importantly, before relaxation, (solid) curves referring to distinct points in the connected geometry follow an ultrafast dynamics causing their overlap. Such behaviour is governed by thermal diffusion and its characteristic time is dictated by electron thermal conductivity and heat capacity. On the contrary, a long-lasting asymmetry of $\Theta_e$ in the disconnected metaatom is observed, namely curves will overlap only when vanishing, \textit{i.e.} when electron excess energy is dissipated. Interestingly, this affects the dynamics of the lattice temperature as well, since its source term (see Eq. (4) from the main text) remains asymmetric over a long time interval. This explains the dynamical behaviour of $\Theta_l$ shown in Fig. S6b, where curves for the disconnected configuration (dashed lines) follow significantly different dynamics and experience a much slower diffusion process (compare thermal conductivities and heat capacities with the electronic gas). Hence, highly asymmetric temperatures entail in turn asymmetric changes in the permittivity (according to the physical processes discussed in Section 3.3), which result in an optical symmetry-breaking window lasting for a much longer time in the disconnected metagrating than in the connected case. Importantly, the dynamics of the transient diffraction management achieved in both structures (see \textit{e.g.} Fig. 5) well correlate with the time evolution of the internal energetic degrees of freedom here presented in the two cases respectively. Indeed, an interplay of $N$ and $\Theta_e$ dominates the response over the first ps, with the contribution arising from thermal hot carriers which affects the symmetry breaking to a lesser degree and a short time interval in the connected structure (Fig. S6b, solid). On the other hand, electron and phonon temperatures govern the signal decay and entail, in the disconnected structure, a long-lasting asymmetry effect.
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