Supplementary Material

**Time-varying Optical Vortices Enabled by Time-Modulated Metasurfaces**

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The Supplementary Material is organized into four sections:

S1. Theoretical Derivation of Topological Charge based on Canonical Momentum Formulation,

S2. Device Simulations and Carrier Dynamics,

S3. Effect of Termination in Biasing Lines on the Performance of the Metasurface.

S4. Extended Discussion on the Efficiency and Mode purity

**S1. Theoretical Derivation of Topological Charge based on Canonical Momentum Formulation**

As it is shown in the manuscript, based on the equivalence between the reflected wave from a TMM with an azimuthal frequency gradient and ideal field profile of a Bessel beam, the topological charge of the dynamic optical vortex due to the $p$-th frequency harmonic can be obtained as $|I_p(z,t)| = p \Delta f \mathcal{N} \left( t + \frac{z}{c} \right)$. Here, in order to provide a more rigorous calculation, we will evaluate the topological charge of the reflected wave directly based on the canonical momentum formulation of light.

For describing the angular momentum (AM) of a light beam, most previous works have taken the advantage of Poynting theorem. However, utilizing this formalism will cause several issues including the incompatibly of the obtained results with quantum optics theory [1],
Abraham-Minkowski dilemma [2] and yielding complex expressions for OAM and SAM [3], [4]. Recently, Bilokh and coworkers proposed a new approach to explore basic properties of OAM, which can overcome the above-mentioned difficulties, [3]. Here, we will briefly discuss the main points of such a formulation; nevertheless, for a more comprehensive discussion, reader can refer to [4]. As it was studied by Bilokh et al., the canonical momentum of light is expressed as

\[ P = \frac{g}{2} \text{Im} \left[ \hat{E}^\ast \cdot \nabla E + \hat{\mu} H^\ast \cdot \nabla H \right] \quad (S1) \]

where \( E \) and \( H \) are electric and magnetic field, respectively while gaussian units with \( g = (8\pi\omega)^{-1} \), \( \hat{E} = \epsilon + \omega d\epsilon/d\omega \), and \( \hat{\mu} = \mu + \omega d\mu/d\omega \) are used. It should be mentioned that we will use the identity \( A.(C)B = \sum_i A_i C B_i \) for the relations \( E^\ast \cdot \nabla E \) and \( H^\ast \cdot \nabla H \) given in Equation (S1) [4]. Moreover, since the background medium is air and it does not possess dispersive behavior, \( \hat{\epsilon} = \epsilon \) and \( \hat{\mu} = \mu \). Therefore, the canonical SAM and OAM densities can be calculated as

\[ S = \frac{g}{2} \text{Im} \left[ \hat{E}^\ast \times E + \hat{\mu} H^\ast \times H \right], \quad \mathbf{L} = \mathbf{r} \times \mathbf{P} \quad (S2) \]

wherein \( \mathbf{r} \) is the position vector. Hence, the spin \( (|\sigma\rangle) \) and topological charge \( (|l\rangle) \) of a light beam can be calculated as

\[ \frac{\mathbf{S}}{W} = \frac{|\sigma\rangle}{\omega k}, \quad \frac{\mathbf{L}}{W} = \frac{|l\rangle}{\omega k} \quad (S3) \]

in which \( W \) is the power density and is defined as \( W = g \left(\hat{E}|E|^2 + \hat{\mu}|H|^2\right)/2 \) and \( \mathbf{k} \) is the wave vector. On the other hand, according to Equation (1) of the main manuscript, the reflected electric field due to the \( p \)-th frequency harmonic generated by the frequency gradient TMM can be expressed as

\[ E_r (r,\varphi,t) = E_0 e_p (r) \exp \left[ i \left( \omega_b + p \omega_n (\varphi) \right) t \right] \exp \left( i \frac{\omega_b + p \omega_n (\varphi)}{c} z \right) \hat{x} \quad (S4) \]

By substituting Equation (S4) into Maxwell’s equations, one can obtain the magnetic field as
Based on Equation (S4) and (S5), the power density will be achieved as 
\[ W = g \omega \varepsilon_0 \varepsilon_a E^2 \varepsilon^2(r). \]
In addition, the expressions of \( \mathbf{E}^* \ldots \nabla \mathbf{E} \) and \( \mathbf{H}^* \ldots \nabla \mathbf{H} \) will be obtained in the cylindrical coordinate system as

\[ \mathbf{E}^* \ldots \nabla \mathbf{E} = E^* \left( \frac{\partial E}{\partial r} \hat{r} + \frac{1}{r} \frac{\partial E}{\partial \phi} \hat{\phi} \right) \Rightarrow \]

\[ \mathbf{E}^* \ldots \nabla \mathbf{E} = E^* \left( \frac{\partial E}{\partial r} \hat{r} + \frac{1}{r} \frac{\partial E}{\partial \phi} \hat{\phi} \right) \Rightarrow \]

\[ \mathbf{E}^* \ldots \nabla \mathbf{H} = H^* \left( \frac{\partial H}{\partial r} \hat{r} + \frac{1}{r} \frac{\partial H}{\partial \phi} \hat{\phi} \right) \Rightarrow \]

\[ \mathbf{E}^* \ldots \nabla \mathbf{H} = H^* \left( \frac{\partial H}{\partial r} \hat{r} + \frac{1}{r} \frac{\partial H}{\partial \phi} \hat{\phi} \right) \Rightarrow \]

\[ \mathbf{H}^* \ldots \nabla \mathbf{H} = H^* \left( \frac{\partial H}{\partial r} \hat{r} + \frac{1}{r} \frac{\partial H}{\partial \phi} \hat{\phi} \right) \Rightarrow \]

\[ \mathbf{H}^* \ldots \nabla \mathbf{H} = H^* \left( \frac{\partial H}{\partial r} \hat{r} + \frac{1}{r} \frac{\partial H}{\partial \phi} \hat{\phi} \right) \Rightarrow \]

Therefore, following the obtained relations, the canonical momentum and SAM density will be attained as

\[ \mathbf{P} = \frac{\delta E^* \varepsilon^2 \varepsilon(r) \left( \frac{t + z}{c} \right) p \frac{d \omega_m(\phi)}{d \phi}}{r}, \quad \mathbf{S} = 0 \]

It should be mentioned that \( \mathbf{S} = 0 \) indicates the spin-free nature of the reflected wave, which abides well with the physics of the problem since the incident wave is a linearly-polarized
Gaussian beam and the TMM does not change the polarization of the light. Hence, according to Equation (S2), the canonical OAM of the reflected light will be achieved as

$$\mathbf{L} = \mathbf{r} \times \mathbf{P} \Rightarrow \mathbf{L} = \frac{geE_0^2e^2(r)}{r}\left(t + \frac{z}{c}\right)p\frac{d\omega_m(\varphi)}{d\varphi} \hat{z}$$  \hspace{1cm} (S8)

which consequently give rises to the topological charge of

$$\frac{L}{W} = \frac{|l|}{k \omega} \Rightarrow |l| = \left(t + \frac{z}{c}\right)p\frac{d\omega_m(\varphi)}{d\varphi}$$ \hspace{1cm} (S9)

As it was comprehensively discussed in the main manuscript, we have discretized the metasurface azimuthally into \(N\) sections each of which are modulated with a frequency of \(f_n(\varphi) = f_{m,0} + \left[N \varphi/2\pi\right] \Delta f\). Therefore, substituting this relation into Equation (S9), one can obtain the topological charge of the \(p\)-th harmonic of the reflected wave with a spatially continuous wavefront as the following while disregarding the discretization-induced errors in the wavefront

$$|l_p(z,t)| = p \Delta f N\left(t + \frac{z}{c}\right)$$ \hspace{1cm} (S10)

Comparing Equation (S10) with Equation (6) of the main manuscript reinforces the validity of the given relation for the topological charge of the dynamic vortices generated by the frequency gradient TMM.

**S2. Device Simulations and Carrier Dynamics**

As it has been thoroughly discussed in the main manuscript, in order realize a TMM for generation of time-varying optical vortices, we have utilized a metasurface consisting of Si/HAOL/ITO/HAOL nanodisk heterostructures located on top of a Si/SiO2/Au stack which renders a dual-gated field effect modulator offering dynamic phase modulation in the reflection mode. In this section, we will provide a more comprehensive discussion about the electrostatic simulations of such a unit-cell in Lumerical Device solver [5] which adopts finite-element method (FEM) to solve the Poisson and drift-diffusion equations while using
Fermi-Dirac statistics. In addition to describing the step-by-step procedure of simulation, we will also outline the dependency of the optical properties of ITO and Si on the applied bias voltage.

For modeling the carrier dynamics within the constituent unit cells of such a metasurface, we have modeled the dual-gated GMRM building blocks with a multilayer structure as shown schematically in Figure S1(a). Two external voltages of $V_1$ and $V_2$ are applied between the ITO and top/bottom doped Si layers from the side with the aid of three external aluminum (Al) electrodes making ohmic contacts with these layers. It should be mentioned that in this work the applied bias voltages are considered to be equal in both magnitude and polarity, that is $V_1 = V_2$.

We have considered both ITO and top/bottom Si layers to be as n-type doped materials with the background carrier concentrations of $N_b^{(\text{ITO})} = 3 \times 10^{20}$ cm$^{-3}$ and $N_b^{(\text{Si})} = 1 \times 10^{19}$ cm$^{-3}$, respectively. ITO is modeled as a semiconductor with electron affinity of $\chi_{\text{ITO}} = 5$ eV [6], electron mobility of $\mu_n^{\text{ITO}} = 25$ cm$^2$ V$^{-1}$ S$^{-1}$, electron effective mass of $(m^*_n)_{\text{ITO}} = 0.35 \times m_0$ ($m_0$ being the electron rest mass and has the value of $m_0 = 9.11 \times 10^{-31}$ kg) [7], DC permittivity of $\varepsilon_{\text{DC}}^{\text{ITO}} = 9.3$ [8] and bandgap energy of $E_{\text{bg}}^{\text{ITO}} = 2.8$ eV [9]. It is notable to mention that as ITO is considered as a degenerately n-type doped material, its corresponding holes do not contribute to the carrier dynamics. This will give us a degree of freedom that to set the hole effective mass and mobility of ITO to be $(m^*_p)_{\text{ITO}} = m_0$ and $\mu_p^{\text{ITO}} = 1$ cm$^2$ V$^{-1}$ S$^{-1}$, respectively without the loss of generality [7].

Similar sets of parameters are defined for the top/bottom Si layers which are given as follows: electron affinity of $\chi_{\text{Si}} = 4.01$ eV [10], DC permittivity of $\varepsilon_{\text{DC}}^{\text{Si}} = 11.7$ [11], bandgap energy of $E_{\text{bg}}^{\text{Si}} = 1.11$ eV [12],[13], electron and hole mobilities of $(\mu_n)_{\text{Si}} = 80$ cm$^2$ V$^{-1}$ S$^{-1}$,
\( (\mu_p)_\text{Si} = 60 \text{ cm}^2 \text{ V}^{-1} \text{ S}^{-1} \) [14], respectively and electron and hole effective masses of \( (m^*_N)_\text{Si} = 1.08 \times m_0 \) and \( (m^*_p)_\text{Si} = 0.81 \times m_0 \). In addition to the ITO and Si layers, the DC permittivity of the HAOL, serving as the gate-dielectric, is selected to be \( \varepsilon_{\text{HAOL}}^{\text{DC}} = 22 \) according to the experimental measurements that has been conducted in Ref [7].

Figure S1. (a) Schematic of the multilayer structure used for electrostatic simulations. Three external Al electrodes are used for applying two sets of bias voltages and making ohmic contacts with the top/bottom n-type doped Si and ITO layers from the side. The electron carrier concentration within the (b) top and (c) bottom ITO active layers as functions of applied bias voltage and distance from ITO-HAOL interfaces. The electron carrier concentration within the active layer of top Si when the applied bias voltage is (d) negative and (e) positive as functions of applied bias voltage and distance from Si-
HAOL interface. (f) The hole density at the Si active layer as functions of applied voltage and distance from Si-HAOL interface. (g)-(i) show the same as (d)-(f) in the bottom Si layer.

Furthermore, it is well known in the physics of solid state devices that work function, which is defined as the required energy to remove one electron from a specific material, of an n-type doped semiconductor has a direct relation with its doping level, affinity and bandgap energy as [15]

\[
\Phi = \chi + \frac{E_{\text{bg}}}{2} - k_B T \ln \frac{N}{N_i}
\]  

whence \( N_i \) is the intrinsic doping of the material, \( k_B = 1.38 \times 10^{-23} \text{ m}^2\text{kg s}^{-2}\text{K}^{-1} \) is the Boltzmann constant and \( T \) is the background medium temperature. At the room temperature where \( T = 300 \text{ K} \) and \( k_B T = 25.7 \text{ meV} \), substituting the above-mentioned parameters into Equation (S11), will give rise to the intrinsic work functions of ITO and Si layers as \( \phi_{\text{ITO}} = 6.4 \text{ eV} \) and \( \phi_{\text{Si}} = 4.56 \text{ eV} \), respectively. Moreover, the work functions of the gold (Au) substrate and the external aluminum electrodes are considered as \( \phi_{\text{Au}} = 5.1 \text{ eV} \) and \( \phi_{\text{Al}} = 4.28 \text{ eV} \), respectively.

In order to be able to accurately resolve spatial distribution of carrier concentrations at the top and bottom interfaces of ITO-HAOL and Si-HAOL interfaces, we have adopted an adaptive meshing with the minimum mesh size of 0.005 nm at the interfaces of semiconductors with gate dielectrics. The carrier distributions of ITO and Si layers are obtained from the Lumerical electrostatic simulation and are brought in Figure S1(b-f) as functions of applied bias voltages and spatial position, which is defined as the distance of each material (i.e., ITO or Si) from its interface with HAOL gate dielectric.

Our electrostatic simulations indicate that HAOL gate dielectric layers with breakdown field of \( E = 7.2 \text{ MV/cm} \) undergo breakdown at the applied bias voltage of 22.5 V. As it is shown in Figure S1(b), increasing the applied bias voltage from -15 V to 22 V will cause the electrons to be accumulated at the top interface of ITO-HAOL before the breakdown of HAOL gate dielectric.
dielectric layers. On the other hand, decreasing the bias voltage towards negative voltages lead to depletion of electrons at the ITO-HAOL interface as it can be clearly seen from Figure S1(b). It is notable to mention that in both cases of accumulation and depletion; the carrier concentration shows an exponentially decaying spatial profile with a screening length of $\approx 3$ nm. Since both top and bottom Si are considered to have the same background carrier concentration yielding the same work function, there is no asymmetry induced by band bending between top and bottom ITO-HAOL interfaces and a similar distribution of carrier concentration can be observed at the bottom ITO-HAOL interface, which is depicted in Figure S1 (c) as functions of applied bias voltage and spatial position.

Meanwhile, in the top Si layer increasing the applied bias voltage from -15 V to 0 V will cause the electrons to be accumulated at the Si-HAOL interface in order to satisfy the charge conservation. However, when the applied bias voltage becomes positive, two regimes in the carrier concentration of top Si layer can be identified as shown in Figure S1 (e) which are demarcated by a threshold voltage of $V_T \approx 4$ V marked by the white dashed line. For applied bias voltage below the threshold voltage, the electrons are depleted at the interface of Si-HAOL in accordance with charge conservation, whole the depletion length increases by increment in the applied bias voltage. Increasing applied bias voltage beyond the threshold voltage, while the length of depletion layer remains almost constant, the holes start to accumulate at the Si-HAOL interface. This in turn will lead to the inversion of the Si surface conductivity from n-type to p-type in this region. The carrier concentration of Si exhibits an exponentially decaying profile as a function of distance from Si-HAOL interface while the screening lengths for electron accumulation, electron depletion and hole accumulation are $\approx 3$ nm, $\approx 20$ nm, and $\approx 1.5$ nm, respectively. Due to the same background carrier concentration in the bottom Si layer and the equal applied bias voltage, all the above-mentioned discussions are also valid for the bottom Si layer as the results of carrier concentrations in this layer are shown in Figure S1 (g)-(i).
By linking the carrier dynamics within the active regions of the designed building block, to the optical response with the aid of carrier-dependent dispersion models of ITO and Si, one can obtain the electro-optical response of the metasurface. To this aim, we have evaluated the changes in silicon permittivity by utilizing Plasma-Drude dispersion model as

$$
\varepsilon_{\text{doped-Si}}(\omega) = \varepsilon_{\text{undoped-Si}}(\omega) - \frac{\varepsilon_e^2}{\varepsilon_0 \omega} \left( \frac{N_{\text{Si}}}{m_{\text{e}*} \omega + i e/\mu_N} + \frac{P_{\text{Si}}}{m_{\text{p}*} \omega + i e/\mu_p} \right)
$$

(S12)

where $m_{\text{e}*} = 0.27 m_0$ and $m_{\text{p}*} = 0.39 m_0$ are the conductivity effective mass of electrons and holes, respectively and $\varepsilon_{\text{undoped-Si}}(\omega)$ is the dispersive permittivity of undoped Si that is taken from the Palik's handbook [16].

By substituting the voltage-dependent spatial distribution of carrier concentrations within Si layers into Equation (S12), the spatial profile of the Si permittivity will be obtained as a function of applied bias voltage in both Si layer at the operating wavelength of $\lambda_0 = 1.552 \mu\text{m}$ which are shown in Figure S2. The stronger variations of permittivity within a distance of $\approx 2$ nm away from top/bottom Si-HAOL interfaces are demonstrated in the inset of each figure.

As can be seen from Figure S2, when the applied bias voltage is negative, the real and imaginary parts of Si permittivity decrease and increase, respectively within a region of $\approx 3$ nm away from Si-HAOL interfaces due to accumulation of electron density. On the other hand, under positive applied bias voltages, the real part of Si permittivity increases, and its imaginary part decreases within a region of $\approx 20$ nm away from Si-HAOL interfaces due to the depletion of electron concentration, prior to reaching the threshold voltage. Beyond the threshold voltage, increasing the applied bias voltage leads to decrement in the real part and increment in the imaginary part of Si within a region of $\approx 1.5$ nm away from the Si-HAOL interface as a result of inversion of Si and accumulation of hole density.
Figure S2. The (a) real and (b) imaginary part of permittivity in the top Si layer obtained from the Plasma-Drude model given in Equation (S12) as functions of applied bias voltage and distance from Si-HAOL interface, at the operating wavelength of $\lambda_0 = 1.552 \mu m$. (c) and (d) shows the same in the bottom Si layer. The stronger variations of permittivity within a distance of 2 nm away from Si-HAOL interfaces are shown in the insets.

In addition to Si layers, the variation in the complex dielectric permittivity of the ITO layer under applied bias voltage is analyzed by considering a Drude model as [7]

$$\varepsilon_{\text{ITO}}(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i \omega \Gamma}$$  \hspace{1cm} (S13)

where $\Gamma = 180 \text{THz}$ is the collision frequency and $\varepsilon_\infty = 3.9$ is the high-frequency permittivity [7] and $\omega_p$ is the plasma frequency which has a direct relation with the carrier density within the ITO layer (i.e., $N_{\text{ITO}}$) as [7]

$$\omega_p = \sqrt{N_{\text{ITO}}e^2/\varepsilon_0 m_{\text{ITO}}^*}$$  \hspace{1cm} (S14)
wherein $m_{\text{ITO}}^*$ is the electron effective mass of ITO, $\varepsilon_0$ is the vacuum permittivity and $e$ is the electron charge. The real and imaginary parts of the permittivity within the ITO layer can be then calculated at the top/bottom ITO-HAOL interfaces by using the voltage-dependent spatial distribution of carrier concentration and are shown in Figure S3 as functions of applied bias voltage and distance from ITO-HAOL interfaces, at the operating wavelength of $\lambda_0 = 1.552 \mu \text{m}$.

Figure S3. The real part of ITO permittivity within the (a) top and (b) bottom active layers at the operating wavelength of $\lambda_0 = 1.552 \mu \text{m}$ as functions of applied bias voltage and distance from ITO-HAOL interfaces. The dashed lines in (a) and (b) denote the ENZ regions formed within the active layers. (c) and (d) show the same for imaginary part of ITO permittivity.

As it can be observed in Figure S3, decreasing the applied bias voltage towards negative values will yield an increment in the real part and a decrement in the imaginary part of ITO
permittivity, which are attributed to the depletion of electrons at the top and bottom ITO-HAOL interfaces. On the other hand, when the applied bias voltage becomes positive, the real part of ITO permittivity decreases and its imaginary part increases by increasing the voltage, as a result of accumulation of electron density at ITO-HAOL interfaces. The variation in the permittivity of ITO under positive applied bias voltages gives rise to formation of epsilon-near-zero (ENZ) regions (i.e., $-1 < \varepsilon_{\text{ITO}} < 1$) at ITO-HAOL interfaces which leads to a significant enhancement of light-matter interaction at nanoscale. Formation of ENZ regions has a crucial role in attaining critical coupling through strong localization of the electric field and driving the resonance from over-coupled to under-coupled state.

In the proposed unit cell, the real part of ITO permittivity crosses zero at the applied bias voltage of $\approx 5$ V (corresponds to $\varepsilon_{\text{ITO}} = 0 + 0.61i$) which coincides with the voltage corresponding to the critical coupling. Moreover, by increasing the bias voltage beyond this point, the direction of the spectral shift in the resonance will be reversed because of the formation of plasmonic regions within the active layers of ITO.

As the final point, we should emphasize that the carrier-induced inhomogenous permittivity profiles of ITO and Si active layers are rigorously taken into account in RCWA simulations of the unit cell through fine discretization of active regions into multiple deeply subwavelength homogeneous layers. For this purpose, we have divided each of the top and bottom ITO active layers with screening lengths of $d = 3$ nm into 15 sub-layers with thickness of 0.2 nm. Moreover, in order to accurately resolve the inhomogeneous profile of permittivity of Si active layer with multiscale screening lengths in different regimes while maintaining the efficiency, we have adopted a nonuniform discretization. That is the first 2 nm of active layer with stronger variations of permittivity (due to electron accumulation and Si inversion) are divided into 10 sub-layers with a thickness of 0.2 nm and the remaining 18 nm of active layer
with smoother variation of permittivity (due to electron depletion) are discretized into 5 layers, each with the thickness of 5.6 nm.

**S3. Effect of Termination in Biasing Lines on the Performance of the Metasurface**

As it has been discussed in the manuscript, in order to independently apply a different modulation frequency to each azimuthal section of the TMM, one must terminate the biasing lines of the unit cells located at the edges of each section. However, it is expected for such a termination to affect the local phase modulation and frequency conversion performance of the designed building blocks, which subsequently alters the performance of the metasurface in generating dynamic optical vortices. In this section, we aim at exploring the effect of biasing line termination on the modulation performance of building blocks and its effect on the overall performance of TMM.

For this purpose, we have introduced a gap in the biasing lines connecting the elements along both $x$ and $y$ directions as shown in Figure S4 (a) which is a representative of the worst-case scenario in terms of perturbation of the optical response. The parameter (Gap) denotes the edge-to-edge distance between the biasing lines. It should be mentioned that for the elements located within the interior part of an angular section $\text{Gap} = 0 \text{nm}$ since the nanodisk heterostructures are interconnected. On the other hand, this parameter is non-zero for the elements located on the edge of an azimuthal section in order to electrically isolate each section from its adjacent ones and enable independent biasing of each section with different modulation frequencies. For analyzing the impact of Gap on the functionality of the metasurface, the first step is to study the effect of this parameter on the performance of the proposed unit cell in terms of its influence on the voltage-dependent phase and amplitude of the reflection. We have limited ourselves to the case of pure frequency mixing which was achieved by employing a sawtooth-like modulation waveform. Similar conclusions can be
drawn for other modulation waveforms giving rise to different spectral diversity of frequency harmonics. The metasurface is under illumination of a normally incident plane wave with transverse electric (TE) polarization at the operating wavelength of $\lambda = 1.552 \mu m$. It is noteworthy to mention that the closer the gap size is to zero, the less change is expected on the functionality of the designed building block. Nevertheless, from the fabrication perspective, this distance cannot approach to near-zero values. Therefore, we have considered the values of Gap that are within the reach of nanofabrication technology. Specifically, we have considered six different values as $\text{Gap} = [0, 10, 15, 20, 25, 30] \text{ nm}$ and evaluated their influence on the amplitude and phase of the reflection. The reflection amplitude and phase are plotted in Figure S4 (b) and (c) as a function of applied bias voltage for different values of the Gap.

**Figure S4.** (a) The schematic depiction of the designed unit cell where a gap is introduced a gap in the biasing lines connecting the elements along both $x$ and $y$ directions. The (b) amplitude and (c) phase of the unit cell at the operating wavelength of $\lambda_0 = 1.552 \mu m$ as functions of applied bias voltage, corresponding to different gap sizes. The normalized conversion efficiency of the time-modulated
metasurface to different frequency harmonics biased with the sawtooth-like modulation waveform optimized for pure frequency mixing with Gap = 0 nm, corresponding to gap sizes of (d) Gap = 0 nm (e) Gap = 10 nm (f) Gap = 15 nm (g) Gap = 20 nm (h) Gap = 25 nm (i) Gap = 30 nm. The stars mark the desired up-modulated frequency harmonics.

As it is shown in Figure S4 (b) and (c), increasing the value of Gap leads to a drastic reduction in the dynamic phase span and a considerable change in the amplitude variations at the operating wavelength. In fact the obtained phase span for the gap values of Gap = 0 nm, Gap = 10 nm, Gap = 15 nm, Gap = 20 nm, Gap = 25 nm and Gap = 30 nm, are 273°, 188°, 126°, 81°, 56° and 43° respectively. The dramatic variation of the phase span with respect to varying the gap size is attributed to the spectral shift of the resonance and the change in its quality factor such that the maximal phase swing resulting from the transition between over-coupled and under-coupled states cannot be attained at the operating wavelength. In the next step, we will evaluate the influence of Gap on the frequency conversion performance of the designed unit cell while adopting the same sawtooth-like modulation waveform which has been optimized for pure frequency mixing with Gap = 0 nm. The normalized conversion efficiency to the frequency harmonics at the operating wavelength of $\lambda_0 = 1.552 \mu m$ for different gap values of 0 nm, 10 nm, 15 nm, 20 nm, 25 nm and 30 nm are depicted in Figure S4 (d)-(i), respectively. As it can be seen from the results, the frequency conversion efficiency to the first up-modulated frequency harmonic drops significantly by increasing the gap size which is mainly attributed to the suppression of dynamic phase span. Moreover, the variation of voltage-dependency of the reflection phase by changing the gap size yields a non-optimal conversion performance by employing the modulation waveform optimized for Gap = 0 nm. In particular, for gap sizes larger than 10 nm, most of the power will be residing at the fundamental frequency rather than the desired
up-modulated frequencies marked by stars. The study shows the gap size of 10 nm to have minimal impact on the performance.

In order to gain more insight into the effect of biasing line termination on the optical response of the metasurface, we have also evaluated the nearfield distributions of electric and magnetic fields within the unit cells in the \((x-z)\) plane for the unbiased case when the gaps are introduced in the biasing lines and the results are shown in **Figure S5**.

**Figure S5.** The electric field distributions within the designed unit cell at the operating wavelength of \(\lambda_0 = 1.552 \mu \text{m}\) for different gap sizes of (a) Gap = 0 nm (b) Gap = 10 nm (c) Gap = 15 nm (d) Gap = 20 nm (e) Gap = 25 nm (f) Gap = 30 nm in the biasing lines. (g)-(l) Their corresponding magnetic field distributions.

The nearfield profiles of electric field in Figure S5 (a)-(f) clearly show that while the electric field is localized at HAOL-ITO-HAOL grating region, allowing for the tunability of light-matter interaction at nanoscale by formation of charge accumulation/depletion layers at the top and bottom ITO-HAOL interfaces under the applied bias voltage, it becomes more
confined in the nanogaps formed by the termination of biasing lines as gap distance increases. The more electric field becomes confined in this nanogap region, the more field intensity decreases within the ITO layer. Such a decrement in the field intensity, yields a narrower tuning range for the optical response and weaker interaction with active region of ITO. In addition to the electric field distributions, the corresponding magnetic field profiles of the given cases are also depicted in Figure S5 (a)-(l). All the nearfield distributions demonstrate a standing wave pattern within silicon guiding core and nanodisks in the transverse direction pointing toward excitation of counterpropagating guided waves which is a manifestation of guided mode resonance. Nevertheless, intensity of the magnetic field within the unit cell hetrostructure decreases by increasing the gap size, which indicates the weaker coupling to the guided modes as a result of spectral position of the resonance by increasing the gap size. Having evaluated the effect of biasing line termination on the constituent unit cells of the metasurface, we will turn into studying its effect on the overall performance of the frequency gradient TMM in generation of dynamic optical vortices. To this aim, similar to the procedure in the manuscript, we have descritized a finite metasurface array consisting of 31×31 unit cells into 16 azimuthal sections and apply a different modulation frequency of 

\[ f_m(\varphi) = N(\varphi/2\pi)\Delta f \]

to each section. The metasurface is illuminated by a normally incident Guassian beam with a waist size of \( w = 13\Lambda \). Since the elements located at the edge of each azimuthal section with introduced nanogaps in the biasing lines connecting them to the adjacent elements are surrounded by interconnected elements, and due to the unaltered nearfield mode profile of the unit cells upon introducing nanogaps, the local response at the edge of azimuthal sections can be described by their periodic response according to local periodicity. As such, we have obtained the overall optical response of the metasurface while taking into account the local response of the elements located on the edge of azimuthal sections in calculation of surface currents by considering the response of unit cells with
different gap sizes in the biasing line. **Figure S6** shows the normalized spatial mode purity of the reflected light from the frequency gradient TMM as functions of OAM state and time for different gap sizes in the biasing line of elements sitting on the edge of azimuthal sections.

**Figure S6.** The normalized spatial mode purity of the reflected light as functions of OAM state and time in the steady-state when the introduced gap parameter in the elements located on the edge of azimuthal section is (a) Gap = 0 nm  (b) Gap = 10 nm  (c) Gap = 15 nm  (d) Gap = 20 nm  (e) Gap = 25 nm  (f) Gap = 30 nm . The dashed lines denote the trajectories corresponding to the dynamic optical vortex generated by the metasurface.

The results point toward generation of a dynamic optical vortex whose topological charge is periodically varying in time within the range of $-8 \leq l(t) \leq 8$ with a linear dependence on time at the intervals of $-0.5 \text{ ns} \leq t \leq 0.5 \text{ ns}$. This beam is a result of dynamic coherent path of generated frequency harmonics of first-order with different frequencies in space-time these dynamic beams are shown by the dashed line in Figure S6. Nevertheless, increasing the gap size gives rise to the emergence of an additional static time-invariant beam with an OAM state of $|0\rangle$ (i.e. a Gaussian beam) which is attributed to the static coherent path of fundamental frequency harmonic in space-time. Particularly, the static beam becomes
dominant in the reflected light beam for gap sizes larger than 10 nm, consistent with the spectral diversity of harmonics shown in Figure S4(d)-(i).

The adverse effects of perturbation in the optical response due to termination of biasing lines at the edge of azimuthal sections can be compensated significantly by enlarging the metasurface aperture due to increase in the ratio of interconnected elements within the interior azimuthal section over the disconnected elements sitting on the edge of azimuthal sections. To demonstrate this, we have performed two simulations in which the size of the metasurface array is increased from $31 \times 31$ elements to $61 \times 61$ and $81 \times 81$ elements, while the value of the introduced gap size in the biasing lines is considered as $\text{Gap} = 10 \text{ nm}$. The same as for the previous cases, each metasurface is discretized into 16 azimuthal sections, which dictates that the obtained topological charge changes in the range of $-8 \leq l(r) \leq +8$ as the time evolves.

The normalized spatial mode purity of the metasurfaces are shown in Figure S7 and compared with those corresponding to $31 \times 31$ metasurface with $\text{Gap} = 0 \text{ nm}$ and $\text{Gap} = 10 \text{ nm}$. As it can be clearly seen from the results, increasing the size of metasurface aperture leads to suppression of the static beam with the OAM state of $|0\rangle$ and decreases the adverse effect of local perturbation in the overall response. In particular, the performance of $81 \times 81$ metasurface with $\text{Gap} = 10 \text{ nm}$ is similar to that of $31 \times 31$ metasurface with $\text{Gap} = 0 \text{ nm}$ in terms of the spatial mode purity of the dynamic optical vortex.

As such, it can be concluded from the study in this section that for a sufficiently large metasurface the effect of biasing terminations for individual biasing of azimuthal sections is negligible on the overall performance.
Figure S7. The normalized spatial mode purity of the metasurface corresponding to (a) 31×31 elements with Gap = 0 nm, (b) 31×31 elements with Gap = 10 nm (c) 61×61 elements with Gap = 10 nm (d) 81×81 elements with Gap = 10 nm.

S4. Extended Discussion on the Efficiency and Mode Purity

The efficiency of a time-modulated metasurface adopting the proposed concept based on azimuthal frequency gradient for time-varying vortex generation can be characterized by two factors: i) the reflection efficiency of the metasurface, and ii) the normalized spatial mode purity of the vortex at each instant of time. While the former is determined by the dissipative loss for reflection, the latter is affected by the efficiency of frequency conversion and azimuthal discretization of the metasurface. The frequency conversion efficiency itself depends on the dynamic phase span and amplitude variations across the phase modulation range in the quasi-static response. The reflection and frequency conversion efficiencies of the time-modulated metasurface considered in this work have been quantified in section 3.2 of the
main manuscript. In this section, we provide further information on the normalized spatial mode purity of time-varying optical vortices and discuss the contributing factors to the perturbation and insufficient mode purity of the total time-domain scattered fields. As mentioned in the main manuscript, the normalized spatial mode purity, can be calculated as

\[ M_{l_i}^{(t)} = \frac{|e_{l_i}^{(t)}|^2}{\sum_j |e_{l_i}^{(t)}|^2} \]  

(S15)

wherein

\[ e_{l_i}^{(t)} = \frac{\int \int_{0}^{2\pi} E(t, r, \varphi) E_{l_i}^{*}(r, \varphi, t) \, dr \, d\varphi}{\int \int_{0}^{2\pi} E_{l_i}^{*}(r, \varphi, t) E_{l_i}^{*}(r, \varphi, t) \, dr \, d\varphi} \]  

(S16)

in which \( E(t, r, \varphi) \) is the reflected field profile at the time instant of \( t \) obtained from modeling the time-modulated metasurface azimuthally divided into \( N \) sections and \( E_{l_i}(t, r, \varphi) \) is the ideal profile of a Bessel beam. By utilizing the above equations, the normalized spatial mode purity of the time-domain reflected field is obtained at time instances corresponding to integer (non-fractional) topological charges \( (l) \) in one cycle of period (i.e., \(-N/2 \leq |l| \leq N/2\)), for three given cases of the main manuscript (i.e., \(-8 \leq |l_{i1}| \leq +8\), \(-8 \leq |l_{i2}| \leq +8\) and \(-4 \leq |l_{i3}| \leq +4\)) and the results are shown in Figure S8. It should be noted that the mode purities are not symmetric in the Hilbert space with respect to \( |l| = 0 \) due to broken azimuthal symmetry of the metasurface (the modulation frequency increases by increasing the azimuthal angle). The considerable reduction of mode purity at the upper and lower bounds of topological charge is due to the periodicity condition and continuity in the OAM states which leads to generation of a superposition OAM state of \(|l\rangle + |-l\rangle\) at \( t = \pm N \Delta f / 2 \). Moreover, the significantly lower mode purities in case of rectangle-like
waveform shown in Figure S8(b) is owing to the superposition state of generated beam at all instants of time.

**Figure S8.** The calculated normalized spatial mode purity of the reflected fields at time instances corresponding to the integer topological charges by employing (a) sawtooth-like wave form for having $-8 \leq |l_{\lambda}| \leq +8$ (b) rectangle-like waveform for achieving $-8 \leq |l_{\lambda}| \leq +8$ and (c) sawtooth-like for generating $-4 \leq |l_{\lambda}| \leq +4$.

The perturbation of nearfield profiles of the time-domain reflected fields (shown in Figure 6 and 7 of the main manuscript) compared to ideal optical vortices stem from two factors: azimuthal discretization of the metasurface and coexistence of parasitic beams in the time-domain field due to the undesired frequency mixing products. While in general the azimuthal discretization and the discontinuities in spatiotemporal phase leads to reduced mode purity at higher topological charges due to more rapid azimuthal variations in the phase, the profile of a vortex beam is not significantly affected by such discontinuities due to topological robustness and self-healing property. As such, the main contributing factor to the imperfection in the profile of the generated optical vortices is the imperfect frequency conversion performance as a result of limited dynamic phase span and nonuniform variations in the amplitude of quasi-static response. As established in Section 2 of the main manuscript, the time-domain scattered field from a frequency-gradient time-modulated metasurface at the steady-state is a superposition of optical vortices resulting from the coherent path formed by different frequency harmonic orders. The topological charge of the vortex generated by $p$-th frequency
harmonic is given by \( I_p(t) = pN\Delta f t \). This implies that if the frequency conversion is perfect and one can convert all the reflected power into a specific harmonic, (e.g., first up-modulated harmonic \( p = +1 \)), a beam with time-varying topological charge of \( I_{+1}(t) = +N\Delta f t \). In order to reinforce this argument and further clarify the role of frequency conversion performance on the nonidealities in the mode profile of time-varying optical vortices, we have simulated the performance of the metasurface in case of pure frequency mixing with a near-unity conversion efficiency. For this purpose, we have assumed that the unit cell provides a \( 2\pi \) dynamic phase span changing linearly as a function of bias voltage (in the range of -17 V to +22 V) while maintaining a uniform amplitude. Employing an optimized sawtooth waveform as shown in Figure S9 (a) in this case yields a frequency conversion to the first up-modulated frequency harmonic with a near-unity efficiency, i.e. almost all the reflected power resides at the first up-modulated frequency harmonic while the share of other harmonics is negligible. Employing this waveform for biasing the TMM divided into \( N=8 \) azimuthal section while incorporating an azimuthally linear frequency gradient profile, the time-domain reflected field from the metasurface is calculates and its normalized spatial mode purity is plotted in Figure S9 (b) as functions of OAM state and time. As expected, the temporal variations in the spatial mode purity shown in this figure point toward generation of a dominant dynamic vortex whose OAM state varies between -8 and +8 with a linear dependence on time at each cycle. Figure S9(c) compares the mode purities at integer topological charges attained with such ideal frequency conversion against the ones afforded by the realized frequency conversion performance of the actual design. It can be observed that, a significant enhancement in the mode purity is obtained as the high frequency conversion efficiency will eradicate the parasitic fields with undesired topological charges due to undesired frequency mixing products. Figure S9(d) demonstrates the intensity of the time-domain reflected field in the ideal case (near-unity frequency conversion efficiency) across the transverse x-y plane at a
distance of $2\lambda_0$ above the metasurface at different instants of time within temporal periodicity cycle of $-1/2\Delta f \leq t \leq 1/2\Delta f$. Rounded donut-shaped profiles with clear on-axis singularity can be observed in this case at each instant of time showing a significant improvement in the nearfield profile of the optical vortices compared to the results shown in Figure 6(e) of the main manuscript.

Nevertheless, as outlined in section 3, due to limited dynamic phase span of the active metasurface and amplitude variations during phase modulation in the actual design, the frequency conversion efficiency to $p = +1$ in the case of pure frequency mixing is limited to 81% which means 19% of the reflected power is residing at other frequency harmonics given by frequencies $f_0 + pf_m$ ($p \neq +1$). These undesirable mixing products give rise to time-varying vortices which coexist with the dominant dynamic vortex with topological charge of
\[ I_{11}(t) = +N \Delta f \cdot t \] in the total time-domain scattered field. Similarly, for the case of concurrent dual-frequency generation, the frequency conversion efficiency to \( p = \pm 1 \) is limited to 76\%.

In particular, the fundamental frequency harmonic \( (p = 0) \) is not sufficiently suppressed as shown in Figure 5(d). This leads to coexisting of a static beam with \( I_0(t) = 0 \) at all instants of time whose footprint can also be clearly observed in the calculated mode purities shown in Figure 6(i).

References


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