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Fundamental Limits to Electromagnetic Response in Novel Platforms

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Final Report

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14. ABSTRACT
Demonstrated and published three key categories of results: (1) development of a framework for upper bounds to EM response in the presence of noise/other sources of incoherence, generalizing the classical ray-optical 'brightness theorem,' (2) derived upper bounds to the maximal free-space concentration of EN waves, particularly as they are confined below the diffraction limit, and (3) demonstrated large-scale inverse design as a tool for discovering state-of-the-art metasurface structures for applications such as lenses and LIDAR.

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Fundamental Limits to Electromagnetic Response in Novel Platforms
PI: Prof. Owen Miller, Yale University
AFOSR Grant #FA9550-17-1-0093
PO: Dr. Arje Nachman

Noteworthy results (abstract):
(1) Developed a framework for understanding electromagnetic-response bounds in the presence of noise or other sources of incoherence. Published in Optica.

(2) Derived bounds to the maximal free-space concentration of electromagnetic waves. Currently on the arXiv. (Accepted to be published in *Physical Review Applied*.)

(3) Demonstrated the use of large-scale computational "inverse design" for state-of-the-art metasurface structures. (Published in *Optics Express* soon after grant end date.)

List of publications supported by grant:
Dec. 2018 – Nov. 2019
- "Limits to surface-enhanced raman scattering near arbitrary-shape scatterers," J. Michon, M. Benzaouia, W. Yao, **O. D. Miller**, and S. G. Johnson, *Opt. Express* 27, 35189-35202 (Nov. 2019).
- "Scattering concentration bounds: brightness theorems for waves," H. Zhang, C. W. Hsu, and **O. D. Miller**, *Optica* 6, 1321-1327 (Oct. 2019).
- "From solar cells to ocean buoys: Wide-bandwidth limits to absorption by metaparticle arrays," M. Benzaouia, G. Tokic, **O. D. Miller**, D. K. P. Yue, and S. G. Johnson, *Physical Review Applied* 11, 034033 (Mar. 2019).
- "Fundamental limits to near-field optical response over any bandwidth," H. Shim, L. Fan, S. G. Johnson, and **O. D. Miller**, *Physical Review X* 9, 011043 (Mar. 2019).
- "Optimal nanoparticle forces, torques, and illumination fields," Y. Liu, L. Fan, Y. E. Lee, N. X. Fang, S. G. Johnson, and O. D. Miller, *ACS Photonics* 6, 395-402 (Feb. 2019).

Dec. 2017 – Nov. 2018
- "Maximal spontaneous photon emission and energy loss from free electrons," Y. Yang*, A. Massuda, C. Roques-Carmes, S. E. Kooi, T. Christensen, S. G. Johnson, J. D. Joannopoulos, O. D. Miller*, I. Kaminer*, and M. Soljačić, *Nature Physics* 14, 894–899 (Sept. 2018)
- "Active radiative thermal switching with graphene plasmon resonators," O. Ilic, N. H. Thomas, T. Christensen, M. C. Sherrott, M. Soljačić, A. J. Minnich, **O. D. Miller**, and H. A. Atwater, *ACS Nano* 12, 2474–2481 (Mar. 2018)

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Dec. 2016 – Nov. 2017

- "Limits to the optical response of graphene and two-dimensional materials," **O. D. Miller**, O. Ilic, T. Christensen, M. T. H. Reid, H. A. Atwater, J. D. Joannopoulos, M. Soljačić, and S. G. Johnson, *Nano Letters* 17, 5408–5415 (Aug. 2017)
- "Topologically enabled optical nanomotors," O. Ilic, I. Kaminer, B. Zhen, O. D. Miller, H. Buljan, and M. Soljačić, *Science Advances* 3, e1602738 (June 2017)
- "Low-loss plasmonic dielectric nanoresonators," Y. Yang*, **O. D. Miller***, T. Chistensen, J. D. Joannopoulos, and M. Soljačić, *Nano Letters* 17, 3238–3245 (Apr. 2017)

List of submitted publications:

- "Tunable metasurface inverse design for 80% switching efficiencies and 144o angular steering," H. Chung and **O. D. Miller**, *arXiv*:1910.03132 (2019).
- "Maximal free-space concentration of electromagnetic waves," H. Shim, H. Chung, and **O. D. Miller**, *arXiv*:1905.10500 (2019).
- "High-NA achromatic metalenses by inverse design," H. Chung and **O. D. Miller**, *arXiv*:1905.09213 (2019).

Interactions with gov't and industry scientists:

Over the past two years I have collaborated with Dr. Richard Vaia, the Senior Scientist for Emerging Materials Systems at the U.S. Air Force Research Laboratory. He is an expert in nanoparticle synthesis, and his group is developing new methods to control their size, shape, and polydispersity. We are leveraging the bounds I have developed to design and synthesize nanoparticles that approach the upper limits of what is possible, with a manuscript to be submitted soon.

Detailed descriptions of this year's noteworthy results are included from the next page.

Noteworthy results (detailed):

Electromagnetic-response bounds in the presence of noise or other sources of incoherence

The "brightness theorem" states that optical radiance cannot increase in passive ray-optical systems. It is a consequence of a phase-space conservation law for optical étendue, which is a measure of the spatial and angular spread of a bundle of rays and has had a wide-ranging impact: it dictates the upper bounds to solar-energy concentration and fluorescent-photovoltaic efficiency, it is a critical design criterion for projectors and displays, and it undergirds the theory of nonimaging optics. Yet a generalization to electromagnetic radiance is not possible, as coherent wave interference can yield dramatic radiance enhancements. A natural question is whether Maxwell's equations, and more general wave-scattering physics, exhibit related conservation laws.

In this work, we develop analogous conservation laws for power flow through the scattering channels that comprise the bases of linear scattering matrices. By a density-matrix framework more familiar to quantum settings, we derive bounds on power concentration in scattering channels, determined by the coherence of the incident field. The ranks of the density matrices for the incoming and outgoing fields play the role of etendue, and maximal eigenvalues dictate maximum possible power concentration. For the specific case of a purely incoherent excitation of *N* incoming channels, power cannot be concentrated onto fewer than *N* outgoing channels, which in the ray-optical limit simplifies to the classical brightness theorem. In resonant systems described by temporal coupled-mode theory, the number of coupled resonant modes additionally restricts the flow of wave etendue through the system. The bounds require only passivity and apply to nonreciprocal systems.

Perfectly coherent excitations allow for arbitrarily large modal concentration (e.g., through phase-conjugate optics), but the introduction of incoherence incurs restrictions. To describe the coherence of incoming waves, we use a density matrix $\boldsymbol{\rho}_{\text{in}}$ that is the ensemble average ($\langle\cdot\rangle$, over the source of incoherence) of the outer product of the incoming wave amplitudes $\boldsymbol{c}_{\text{in}}$:

$$\boldsymbol{\rho}_{\text{in}} = \langle \boldsymbol{c}_{\text{in}} \boldsymbol{c}_{\text{in}}^{\dagger} \rangle.$$

The incoherence of the outgoing channels is represented in the corresponding outgoing-wave density matrix,

$$\boldsymbol{\rho}_{\text{out}} = \langle \boldsymbol{c}_{\text{out}} \boldsymbol{c}_{\text{out}}^{\dagger} \rangle.$$

The density matrices $\boldsymbol{\rho}_{\text{out}}$ and $\boldsymbol{\rho}_{\text{out}}$ are Hermitian and positive semidefinite.

Through passivity and the construction of optimal coherent-wave solutions, we can show that for any level of incoherence, there is a bound on the ensemble-averaged power flowing through any state $\hat{\boldsymbol{u}}$:

$$\langle \left| \boldsymbol{c}_{\text{out},\hat{\boldsymbol{u}}} \right|^2 \rangle \leq \lambda_{\max}(\boldsymbol{\rho}_{\text{in}}) \,.$$

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This expression is a key theoretical result of our work. It states that for a system whose incoming power flow and coherence are described by a density matrix $\boldsymbol{\rho}_{\text{in}}$, the maximum concentration of power is the largest eigenvalue of that density matrix. It ranges from value 1 for a coherent input to $1/N$ for perfectly incoherent inputs over N channels. We prove that for continuous plane-wave channels in homogeneous media, this expression simplifies to the ray-optical brightness theorem.

To probe the channel-concentration bounds, we considered control of diffraction orders through complex metasurfaces for potential applications such as augmented-reality optics and photovoltaic concentrators. Figure 1(a) depicts a designable gradient refractive-index profile with a period of2λ and a thickness of 0.5λ. We consider the bounds for both fully incoherent as well as partially coherent excitations, and we show that inverse-design structures (insets) can approach (but not surpass) the corresponding bounds.

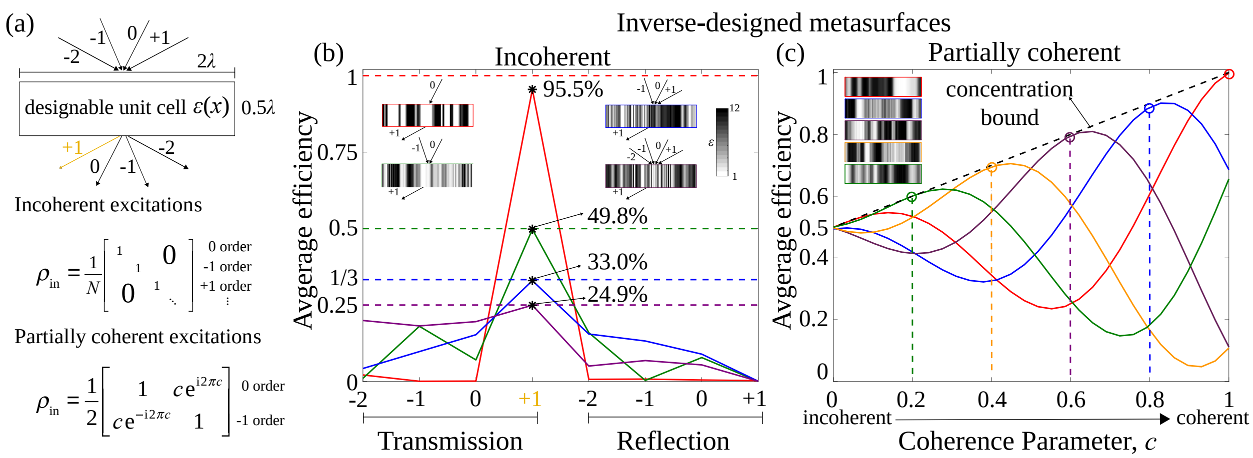

Figure 1. (a) Periodic metasurface element to be designed for maximal power in the +1 transmission diffraction order (yellow). We consider incoherent excitations among the four incident orders, with a diagonal density matrix, as well as partially coherent excitations between the 0 and −1 order, represented by an off-diagonal term with coherence parameter c*. Inverse-designed metasurfaces closely approaching the coherence- and channel-dependent bounds are shown in (b) for incoherent excitations among up to four channels, and in (c) for partially coherent excitations between two channels.*

We further proved in this work that our wave-etendue ideas extend to resonances within the scatterers. For any coupled-mode analytical framework, the number of resonances that are connecting incoming-wave channels to distinct outgoing-wave channels represent another potential bottleneck where energy may have to be lost to satisfy etendue constraints. We provide specific examples of this in waveguide-junction systems, showing the multimode systems can have superior robustness to systems with fewer modes than incoming/outgoing-wave channels.

We are extending this work to a general theory of density-matrix constraints in classical electromagnetism. Our maximum-eigenvalue bound from above can be generalized to any sum of the first M eigenvalues of the density matrix using standard techniques for semidefinite programming. We can use this for a variety of applications; a particularly interesting one is in the free-space concentration of electromagnetic radiation, where it predicts, for example, that it is impossible to confine light to have a suitably defined “width” smaller than the diffraction limit.

Maximum free-space concentration of electromagnetic waves

It *is* possible to confine light to regions smaller than the diffraction limit through engineering the sidebands, i.e., by engineering sub-diffraction-limited "zeros" of the waves but having nonzero amplitude beyond the zeros. A natural question is the bounds on such a process—how much energy must unavoidably arise in the sidebands? What is the maximum energy concentration between the sub-diffraction-limited zeros? We answer these questions in our manuscript "Maximal Free-Space Concentration of Electromagnetic Waves."

We derive upper bounds to free-space concentration of electromagnetic waves, revealing the maximum possible focal-point intensity (related to the well-known ``Strehl ratio'') for a fixed source power and for any desired spot size. For waves incident from any region of space---generated by scattering structures, spatial light modulators, or light sources of arbitrary complexity---we show that the non-convex beam-concentration problem can be transformed to a quadratic program with easily computable global optima. We also extend this approach to derive maximum intensity independent of the exit surface of an incident wave. Our bounds simplify to those derived by Fourier analysis of prolate spheroidal wave functions in the scalar 1D limit. For very small spot sizes *G*, which are most desirable for transformative applications, we show that the focal-point intensity must decrease proportional to G^4, a dimension-independent scaling law that cannot be overcome through any form of wavefront engineering. The bounds have an intuitive interpretation: the ideal field profile at the exit surface of an optical beam-shaping device must have maximum overlap with the fields radiating from a dipole at the origin yet be orthogonal to the fields emanating from a current loop at the spot size radius. We compare theoretical proposals and experimental demonstrations to our bounds, and we find that there is significant opportunity for order-of-magnitude intensity enhancements at those small spot sizes. We use ``inverse design.'' a large-scale computational-optimization technique, to design metasurfaces that generate nearly optimal wavefronts and closely approach our general bounds.

A key theoretical result of our work is a bound on the maximum intensity at the origin in a plane of interest, given the requirement of zero field on a contour C within a sub-diffraction distance of the origin. For free-space Green's-function matrices from the aperture to the origin, $\mathbf{\Gamma}_0$, and from the aperture to the zero-field contour, $\mathbf{\Gamma}_C$, we find a maximum intensity of

$$I \leq \mu^\dagger \left[\mathbf{\Gamma}_0\mathbf{\Gamma}_0^\dagger - \mathbf{\Gamma}_0\mathbf{\Gamma}_C^\dagger\left(\mathbf{\Gamma}_C\mathbf{\Gamma}_C^\dagger\right)^{-1}\mathbf{\Gamma}_C\mathbf{\Gamma}_0^\dagger\right]\mu,$$

where μ is the optimal field polarization at the origin (and is the solution of a small 6x6 eigenproblem). Although it may have an abstract appearance, this expression is a decisive global bound to the optimization problem, requiring only evaluation of known free-space Green's functions.

Figure 2 compares many theoretical designs to our bounds, using the Strehl ratio as the measure of the intensity at the origin and a normalized spot size for comparison across many devices. One can see that at larger spot sizes, close to the diffraction limit, there are designs that can come close to our bounds. At smaller spot sizes, however, the gap between the bounds and current designs is quite large, suggesting there is opportunity for significant improvement over the current state-of-the art.

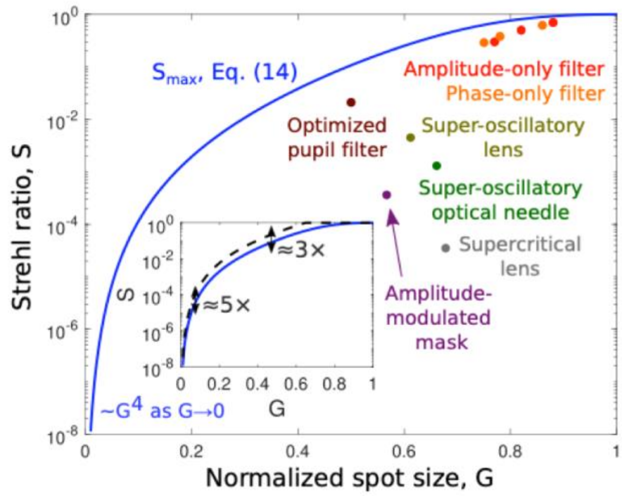

Figure 2. Comparison of global upper bound to Strehl ratio (Eq. 14) to state-of-the-art designs (colored markers). Inset: comparison of our bound (blue line) to the computational bound of Sales & Morris (back dashed), which is 3-5X larger despite applying to a much smaller set of structures (rotationally-symmetric weak scatterers).

One surprising aspect of our work is that our bounds are *smaller* than previous well-known bounds of Sales and Morris, as shown in the inset of Fig. 2, despite our allowance for far more general device structures (strong-scattering, vector-field engineered, non-symmetric, etc.). This validates the robustness of our analytical approach.

Figure 3 represents our designs, which outperform the current state-of-the-art and approach quite close to our bounds. We use large-scale inverse design to discover refractive-index profile that can achieve sub-diffraction-limited focusing with nearly optimal intensity profiles.

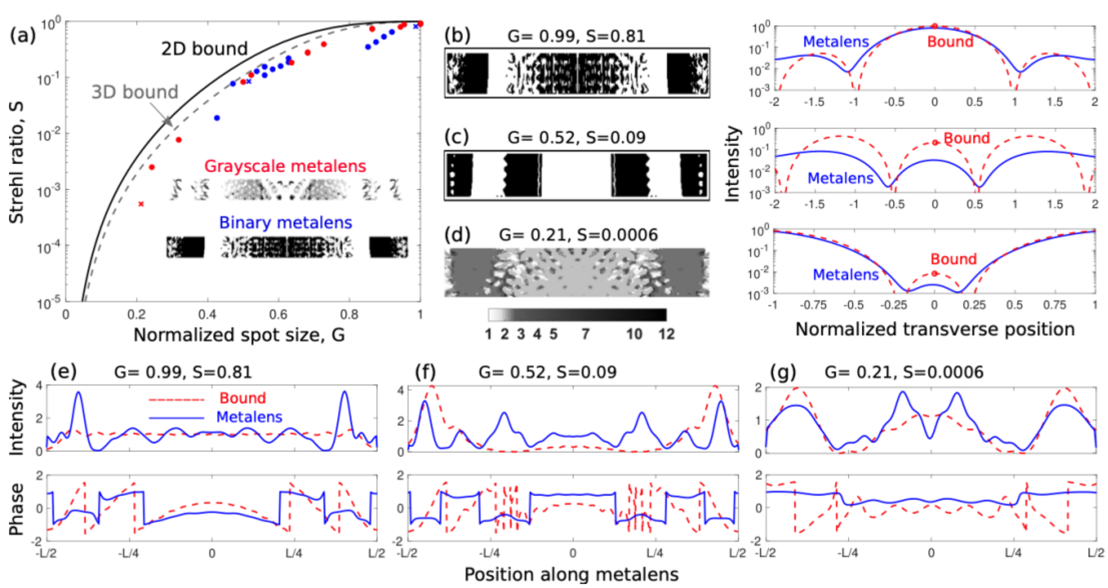

Figure 3. Inverse-designed structures that approach our superresolution bounds, across a large range of spot sizes. (e-g) show the intensity and phase of the fields at the exit planes of the metalenses, alongside the optimal profiles predicted via the bounds.

Inverse Design for State-of-the-Art Metasurfaces

We employed inverse-design techniques to design two classes of metasurfaces that offer state-of-the-art performance: high-NA achromatic metalenses ("High-NA achromatic metalenses by inverse design," *Optics Express* 28, 6945, 2020) and switchable metasurfaces for beam-steering applications ("Tunable metasurface inverse design for 80% switching efficiencies and 144-degree angular steering," arXiv:1910.03132).

Figure 4 summarizes our high-NA achromatic metalens design results, comparing our designs (blue and red curves) to the state-of-the-art designs at the time of publication.

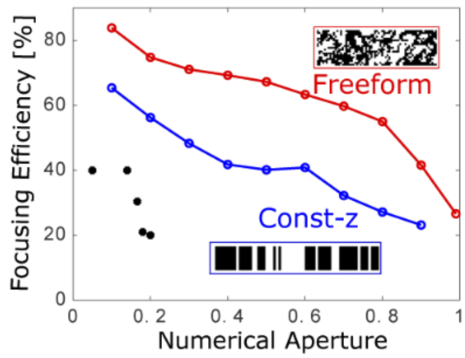

Figure 4. Our designs (const-z and freeform, corresponding to the degrees of freedom in each case) compared with previous literature (black markers).

For every NA our designs achieve higher focusing efficiencies. Figure 5 demonstrates the capability of one of our designs to achieve broadband focusing even at an NA of 0.9.

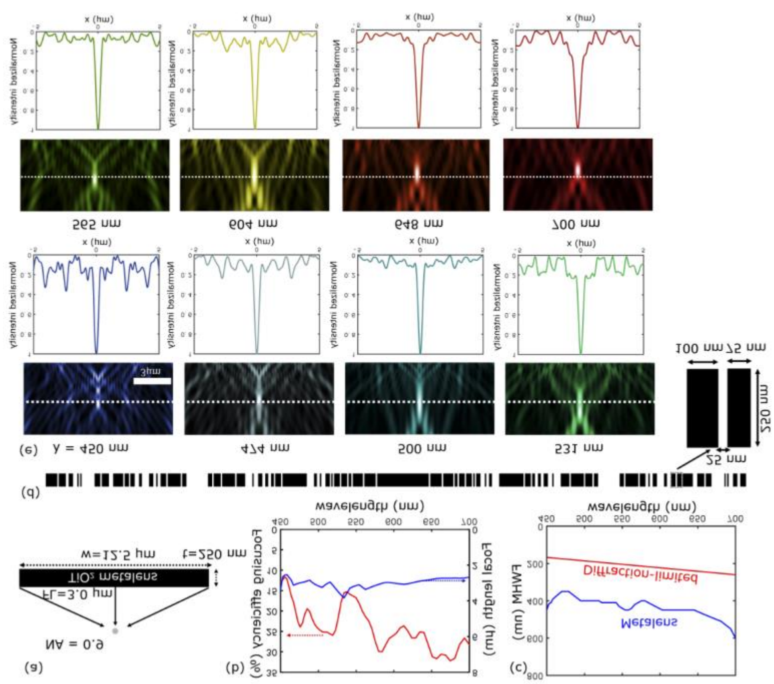

Figure 5. High-NA achromatic metalens design.

We applied the same design principles to the discovery of high-efficiency *switching* metasurfaces with embedded liquid-crystal media. Figure 6 shows a similar comparison of our designs (red, blue, purple solid lines) to the state-of-the-art (back, orange markers).

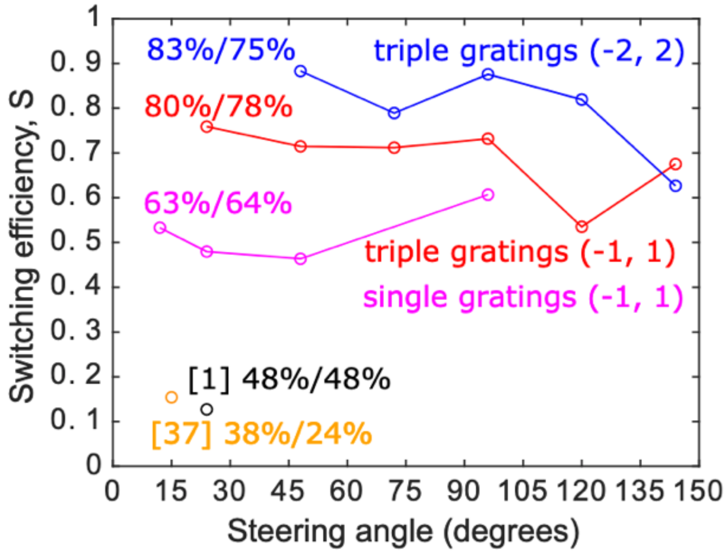

Figure 6. Comparison of our inverse-designed switchable metasurfaces (single and triple gratings) to the state-of-the-art (markers).

Figure 7 shows a specific design, which is able to achieve an unprecedented $\pm 72°$ switching with high fidelity. Simulated with a Gaussian beam, one can clearly see the signal with only a very small amount of noise; the first design in the literature to achieve signal significantly larger than the noise.

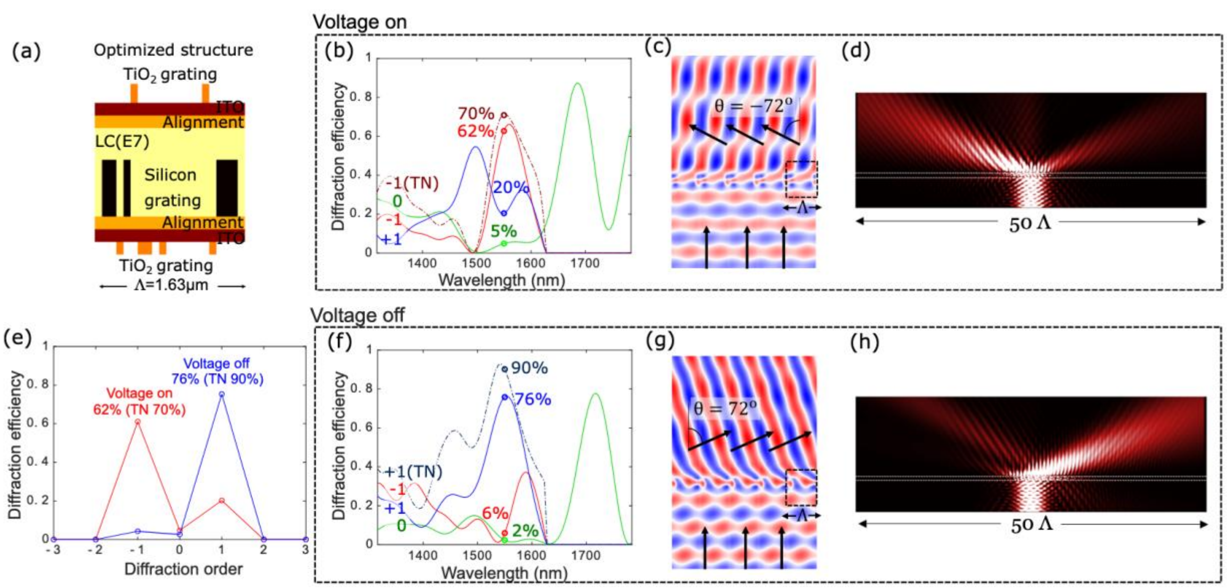

Figure 7. (a) Optimal liquid-crystal structure for 144-degree switching. (b-d) voltage-on characteristics. (f-h) voltage-off characteristics.