Broadband high-efficiency chiral splitters and holograms from dielectric nanoarc metasurfaces

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Abstract

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Simultaneous broadband and high efficiency merits of designer metasurfaces are currently attracting widespread attention in the field of nanophotonics. However, contemporary metasurfaces rarely achieve both advantages simultaneously. For the category of transmissive metadevices, plasmonic or conventional dielectric metasurfaces are viable for either broadband operation with relatively low efficiency or high efficiency at only a selection of wavelength. To overcome this limitation, we propose dielectric nanoarcs as a means to accomplish two advantages. Continuous nanoarcs support different electromagnetic resonant modes at localized areas for generating phase retardation. Meanwhile, the geometric nature of nanoarc curvature endows the nanoarcs with full phase coverage of 0-2π due to the Pancharatnam-Berry phase principle. Experimentally incorporated with the chiral-detour phase principle, we demonstrate a few compelling functionalities, such as chiral beamsplitting, broadband holography, and helicity-selective holography. The continuous nanoarc metasurfaces prevail over plasmonic or dielectric discretized building block strategies and our findings will lead to novel designs of spin-controllable metadevices.

1. Introduction

The ability to manipulate light at nanoscale is the cornerstone for a wide range of applications including ultra-compact optoelectronic and on-chip photonic devices.[1-3] Recently, metasurfaces have been satisfying current requirements for miniaturization of optical systems without compromising their original functionalities and performances. A series of unprecedented developments have been demonstrated, such as invisible cloak,[4] multifunctional metalens,[5, 6] and nanometric hologram.[7] Besides them, control of light through manipulating its spin states is one of the frequently demanded functions. For instance, giant circular dichroism arising from intrinsic chirality has important applications in polarization optics and spintronics.[8, 9] Additionally, chiral beamsplitters, which separate the incident beam into different output directions depending on the helicity (circularly polarized states) of light, enable to encode and process binary information in optical communication.[10-13] Chiral holograms, which reconstruct different images by altering the handedness of incident light, hold great promise in 3D imaging, data storage, beam shaping.[14-16] Although metasurfaces have potential to replace or complement their conventional refractive and diffractive counterparts for the realization of miniaturized nanophotonic devices, contemporary metasurfaces still face the challenge of possessing both broadband and high efficiency merits simultaneously.

The two features of broadband and high efficiency are often related with plasmonic and dielectric metasurfaces. On one hand, the broadband attribute can be generally achieved by plasmonic metasurfaces.[17-19] Considering typical metallic building blocks, e.g., nanoslits or nanorods, behave like broadband polarizers that follow the Malus law of absorption,[20] they easily broaden the operation range.[18] Although plasmonic metasurfaces feature wide bandwidth, the power losses of plasmonic devices are prohibitively high, making the application of transmissive metadevices inefficient. Conversely, on the other hand, dielectric counterparts can overcome the critical issue of high-energy dissipation because light does not couple to plasmons or optical phonons in this case.[2, 23] Nevertheless, most of the previous efforts to design dielectric metasurface relied on a discretized strategy, i.e., numerous nanoantennas are arranged in an array.[24, 25] This strategy is viable over a narrow spectral range. In other words, dielectric metasurfaces, in spite of high efficiency, are always plagued by the shortcoming of limited bandwidth. For improving the bandwidth, the routine methods make use of the complicated super-unit cells which is composed of different morphological (sizes and shapes) building blocks as subunits. Consequently, these involved subunits accordingly operate at different wavelengths
for fulfilling a discretized broadband.[26, 27] In fact, this mechanism of segmented or interleaved subunits originates from the concept of shared-aperture phased array antennas.[28] However, these methods generally lead to the exponential decrease of signal-to-noise ratio with the increase of channel number (multiple wavelengths).[29] Moreover, the issue that discrete nanoantennas weakly response to continuous spectrum remains to be addressed.

The geometric phase or the Pancharatnam–Berry (PB) phase methods prevail over the counterparts employing size-variable resonators from the perspective of dispersionless property,[30, 31] as the fundamental principle of PB phase is only related with wavelength-independent polarization evolution on Poincaré sphere.[32] The advantage endows contemporary metasurfaces with the possibility of expanding the bandwidth. However, the building blocks that produce the PB phase are dispersive because whether polarizer-functional or half-waveplate-functional building blocks inherently valid over a finite spectral range. As a consequence, the combination of PB phase principle and traditionally discretized building blocks is still incapable of both broadband and high efficiency. To surmount this hurdle, we propose dielectric nanoarc metasurfaces working towards broadband and high efficiency simultaneously. Notably, the advantage arising from the novel nanoarc structures is to generate a continuous geometric phase gradient, which is of great interest to chiral beamsplitting and multiplexing hologram. The concept of continuous building blocks has been introduced in several references;[18, 33, 34] however, these reports only focus on the plasmonic metasurfaces without adequate efficiency. In addition, the unit cell of nanoarcs is different from traditional dense gratings.[35-37] By modulating electromagnetic resonance, a single-curved nanoarc structure can give rise to strong chiral light-matter coupling and result in higher efficiency.

In this paper, we propose dielectric nanoarc structures as building blocks that support different electromagnetic resonant modes at localized areas. These dielectric nanoarcs have the distinct advantage of a continuous-phase gradient along the curved trajectory within an individual building block. Of particular interest is the case where a nanoarc is deformed as a half-circle, giving rise to a continuously full-phase (0-2π) coverage. This flexibility in phase control can enable chiral beamsplitting over a broadband spectrum. Furthermore, by incorporating with the principle of chiral-detour phase, we are also able to demonstrate complex holograms of diffracting light with high efficiency into the handedness-dependent diffraction order. The continuous building blocks of nanoarc metasurfaces outperform their discretized counterparts in terms of both the bandwidth and high efficiency and innovate a new class of designs in dielectric metadevices.

2. Results and Discussion

Figure 1a shows the conceptual scheme of the proposed dielectric nanoarcs that response to broadband chiral light, i.e., circularly polarized light. When periodic nanoarcs are formed in a two-dimensional array, they have the ability to steer chiral lights into different diffraction orders, providing the same functionality as a blazed diffraction grating. Left- and right-circularly polarized (LCP and RCP) light beams are discriminated into different paths versus the
symmetric axis whilst impinging on this polarization grating.\textsuperscript{[38]} The phenomenon can also be interpreted by the spin-Hall effect stemming from the PB phase.\textsuperscript{[39]} We assume normal incidence of an electromagnetic wave $E$ with Jones vector components $[E_x, E_y]^T$. In the transmission mode, the vectorial Fourier coefficients and diffraction efficiency of the ±1 diffraction orders are expressed respectively\textsuperscript{[40]}

$$D_{z\pm} \propto (t_{Te} - t_{TM})(E_x \ iE_y) \begin{pmatrix} 1 \\ i \end{pmatrix}$$

$$\eta_{z\pm} \propto |t_{Te} - t_{TM}|^2 \left[ 1 \pm \frac{|E_x||E_y|\sin(\Delta \Phi)}{|E_x|^2 + |E_y|^2} \right]$$

(1) (2)

where $t_{Te}$ and $t_{TM}$ are the complex-amplitude transmission coefficients of the grating for TE- and TM-polarized input waves respectively and $\Delta \Phi$ is the phase difference between the $E_x$ and $E_y$ components. Equation (1) shows that the polarization states of the ±1 diffraction orders are both circularly polarized lights. It can be interpreted from Equation (2) that when circularly polarized lights, i.e, $\Delta \Phi = \pm \frac{\pi}{2}$, impinging on the polarization grating, the diffraction efficiency of the ±1 diffraction orders can be close to 100%. In other words, the zero diffraction order disappears. The prerequisite is that the element modulating the polarization state of incident light function as an ideal half-waveplate. In our case, the element is a continuous nanoarc structure (Figure 1b). In fact, the chirality inversion,\textsuperscript{[41]} also called the spin-flip,\textsuperscript{[42]} actually takes place at localized areas due to different resonant modes relating to a rotating reference frame $(u, v)$ (Figure 1b and 1d). From the viewpoint of an infinitesimal area, the curved nanoarc can be
regarded as a straight-line structure (inset in Figure 1d). We used the finite-difference time-domain (FDTD) method to perform full-field electromagnetic simulations. By examining the scattering cross-section we confirmed that the magnetic dipole resonance is excited along the $u$-polarization illumination.\(^{[43]}\) No notable resonance was observed along the $v$-polarization illumination, which brings about substantial phase retardation (e.g., $\pi$ shift).\(^{[44]}\) Note that we systematically investigated various nanoarc structures, e.g., circle, ellipse, catenary and parabola, to examine their chiral beamsplitting and hologram efficiencies. Eventually, the half-circle nanoarc was selected for the subsequent experiment with the trade-off consideration of geometric phase coverage, phase chiral extinction ratio and absolute efficiency (see supplementary materials). When this nanoarc was deformed as a half-circle, the yielding PB phase continuously varied from $-\pi$ to $\pi$ between the left and right endpoints along the $x$-axis (Figure 1c) because the PB phase has a geometric relation $\Phi = 2\sigma\theta(\xi)$ to the orientation of continuous path $\xi$, where $\sigma = \pm 1$ denotes the RCP and LCP, $\theta(\xi)$ is the inclination angle with respect to $x$ coordinate, explicitly $\theta(\xi) = \tan^{-1}(dy/dx)$. Thus, the observed phenomenon of chiral beamsplitting stems from the geometric nature of anisotropic nanoarcs.

To optimize the transmission characteristic, we systematically investigate the total transmission coefficients and conversion efficiency corresponding to varying structural parameters and wavelengths using the FDTD method (Figure 2). A single half-circle of the hydrogenated amorphous silicon (a-Si:H) as shown in Figure 1b was placed onto a glass substrate and a light source with RCP was illuminated normally from the glass to the nanoarcs. A periodic boundary condition is used in the $x$- and $y$-directions with the lattice constants of 1 $\mu$m and 0.5 $\mu$m, respectively. The thickness $H$ and the width $w$ of the a-Si:H nanoarc were varied to maximize the wavelength range for both high transmission and conversion. The outer radius $R$ of the half-
circle nanoarc was 0.45 μm. The total transmission coefficient and the conversion efficiency are defined as follows: \( T_{-\sigma_-} + T_{-\sigma_+} \) and \( T_{-\sigma_-} / (T_{-\sigma_-} + T_{-\sigma_+}) \). From the simulation results shown, the majority of nanoarcs with different heights exhibit high transmission, which is attributed to the low loss of silicon over this frequency range. The structural width, however, plays a key role in the conversion efficiency. As previously mentioned, the nanoarc with a chosen width supports magnetic dipole resonance excited by longitudinally polarized light, which means that the displacement current loop is enhanced inside the transverse plane of nanoarc.\(^{[45, 46]}\) As the width increases, the resonant mode disappears gradually. The phase difference between the incident and scattered light waves therefore becomes less obvious and the spin conversion efficiency decreases accordingly. By considering the trade-off between the transmission and the conversion efficiencies, we chose a width and height of 90 nm and 190 nm, respectively, for the a-Si:H nanoarc as the building block for subsequent experiments.

In general, the chiral response of light is associated with either intrinsic or extrinsic chirality of molecules. By contrast, the nanoarcs possess mirror-image symmetry. Figure 3a represents a top-view scanning electron micrograph (SEM) of a fabricated nanoarc array. In this case, the phase gradient of \( d\Phi / dx \) is constant. According to the generalized Snell's law,\(^{[47]}\) it is easy to show that the transmission angle is \( \theta \propto \lambda \cdot d\Phi / dx \). The propagation of the wave front of the transmitted \( E_z \)-field of the normal incident RCP light at \( \lambda = 800 \) nm is visualized in Figure 3b to aid an intuitive understanding. The LCP light experiences the same behavior as the RCP light with the opposite transmission angle of \(-\theta\) owing to the symmetry. Figure 3c reveals the numerical simulation of helicity-dependent beamsplitting in the momentum space and the far-field intensity distribution (insets in Figure 3c). The curved chain gives rise to an additional
phase of \( \Phi = \int \Delta k \cdot d \ell \), where \( k \) defines the wave-vector. It is demonstrated that the spin-flip deflection originates from the momentum shift of 
\[
\Delta k = \left| \frac{2 \pi \sigma}{P_x} \right|
\]

To verify the nanoarc performances of broadband and high efficiency, we carried out the far-field experiments with a tunable laser. The details of the experimental setup for chiral beamsplitting are described in the Supporting Information. The overall transmission surpassed 50% at wavelengths longer than 700 nm and reached up to 95% at approximately 800 nm (Figure 3d). The spectral range over which chiral beamsplitting occurs approximately corresponded to the electromagnetic resonance span (Figure 1d). The phenomenon of chiral beamsplitting based on the geometric phase turned out to be closely related to the polarization conversion efficiency, which means that only transmitted light with a cross-polarization state would sustain the helicity-dependent redirection. Because co-polarized light is not able to satisfy the closed-path evolution of polarization over the Poincaré sphere, the maintained polarization component does not accumulate any extra phase for anomalous diffraction. We experimentally measured the diffraction patterns under RCP and LCP illumination at the wavelength of 800 nm (Figure 3e). Because the nanoarcs have a very high spin-flip purity at the central frequency, the zero diffraction order almost disappears in the experiment, even without the help of the cross-polarization strategy in the far-field detection. At other wavelengths, the RCP and LCP lights are also steered into the ±1 diffraction orders (see Supporting Information). These observations verify that chirality dependent redirection can be accomplished by the nanoarc grating.

Since the majority of chiral light can be steered into a desired diffraction order, we have the opportunity to create compelling holograms by rationally disposing the nanoarcs with one
another. Wavefront forming is achieved by implementing the detour-phase principle.[48] Note that the traditional efforts employing the detour-phase concept are mostly devoted to helicity independent holograms.[49,50] As a consequence, the modulated phase definitely results in two isomorphic holograms at the twin diffraction orders, which incurs the expense of a half efficiency. Moreover, the plasmonic nanoslits are only sensitive to linearly polarized (LP) light without chiral discrimination.[51] In our case, we exploit the continuous nanoarcs which are able to deflect the incoming light into different specific directions based on incoming handedness with high efficiency and selectivity. This approach expands the concept of the chiral-detour phase and we also develop the innovative building blocks to demonstrate the versatility of the broadband property. As shown in Figure 4a, the nanoarcs are disposed along the x-axis pixel by pixel, at locations δ₁, δ₂,..., δₜ for selecting the desired phase shifts. We selected the example of a ‘Chinese knot’ pattern to reconstruct by our method (Figure 4b). The iterative Fourier transform algorithm, also called Gerchberg–Saxton algorithm, is employed to determine the phase-only function by

\[ F\{H_p(x,y)\} = A(x,y)e^{-i\varphi(x,y)} \quad (3), \]

where \( H_p(x,y) \) is the phase of hologram, \( A(x,y) \) is the target amplitude distribution, and \( \varphi(x,y) \) is the phase of reconstructed light. In Figure 4c, the phase profile is obtained using the algorithm to generate the target holographic image (Figure 4b). Owing to the limitation of the fabrication area, we used a 200 × 200 pixels picture for a proof of concept demonstration. In fact, the light wavelets from the adjacent nanoarc scatterers were phase-shifted with respect to one another by the relationship of \( \Delta \varphi = 2\pi\delta_\alpha / \lambda \cdot \sin \theta \), which has an intrinsic dispersion within the spectral range whilst the diffraction angle is a function of wavelength as

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\[ \theta_a = \arcsin \left( \frac{\lambda}{P_a} \right) \]

Hence, the phase difference becomes wavelength independent because the dispersion is compensated from the view of the observer, which is advantageous for achieving broadband holograms. See more details in the Supplementary Materials for the hologram experiment. The experimental results are revealed in Figures 4e-h. Although the clarity of the reconstructed images depends on the wavelength, it is demonstrated that the chiral-detour phase holograms operate the target intensity profile at a wide wavelength range from 600 to 900 nm.

According to what has been discussed so far, we concentrate on the capability of continuous nanoarcs for chiral beamsplitting and helicity-dependent holograms. In addition, the vectorial behavior of nanoarc’s route, \( \vec{\zeta}(x,y) \), is taken into consideration for the helicity multiplexing holograms (see more details in Supporting Information). Helicity-selective different holograms are attributed from the interaction between the polarization of incident light and the orientation of nanoarcs. The orientation of a half-circle nanoarc is closely related with the azimuth of diffraction for light of proper handedness light. Switching the nanoarc inversely results in changing the diffraction angle from \( \theta \) to \( -\theta \). We adopted the interleaved array of inverse nanoarcs along the \( y \) axis and encoded different symbols representing the spin state information, i.e., \( \sigma_{+} \) and \( \sigma_{-} \), with two phase-shifted images; see Figure 5. In this way, helicity-selective images are obtained experimentally, depending on the light handedness. Because the linearly polarized light can be viewed as consisting of equal energies of RCP and LCP light, two symbols both appear accordingly.

3. Conclusion
In conclusion, we demonstrate the dielectric nanoarc metasurfaces which operate simultaneously with high efficiency and broadband features. A simple half-circle nanoarc can locally function as the idea half-waveplate in replace of an array of traditional nanoantennas. By delving more deeply into the fundamental mechanism, we find that the nanoarcs support different electromagnetic resonant modes for generating the phase retardation with respect to the orthogonal directions and meanwhile the geometric nature of curved path endows the nanoarcs with a full coverage of 0-2π due to the PB phase principle. Consequently, almost all the incoming energy can be modulated into the chiral dependent transmission orders over a wide spectral range. We have shown that a few compelling functionalities, such as chiral beamsplitting, broadband holograms, and helicity selective holograms, can be achieved in super-thin, small, and lightweight nanoarcs. In addition, combining the continuous nanoarcs with other complex-path configurations, the realization of numerous striking phenomena, e.g., vortex focusing, generating and spin-orbit conversion, can be envisioned. Our findings will also lead to the design of novel spin-controllable nanodevices and facilitate further development of nanophotonics.

4. Experimental Section

Sample preparation: For the hydrogenated amorphous silicon (a-Si:H) deposition, the fabrication of the nanoarc metasurfaces started from cleaning of a slide glass with acetone/IPA/deionized water to promote the adhesion between the substrate and a-Si:H film. Following this, a 190 nm thick a-Si:H was deposited on the substrate by plasma-enhanced chemical vapour deposition (Plasmalab 100 from Oxford) using optimized conditions in the previous work. Ellipsometric characterisation shows that the refractive index of the film is around 4 in 600-900nm range, while the extinction coefficient is negligible beyond 700nm.
**Nanofabrication:** The nanoarc metasurfaces were fabricated by means of electron-beam lithography (Vistec EBPG 5000 Plus ES) on a silicon deposited glass substrate, using poly(methyl methacrylate) (Micro-Chem, 950k A4) and Copolymer (Micro-Chem, EL6) as a bilayer resist stack for a standard lift-off process and a sacrificial chromium layer (20 nm) for the charge dissipation during the exposure. The structures were developed with a 1:3 (by volume) mixture of methyl isobutyl ketone/2-propanol for 90 s, rinsed with 2-propanol, and dried with a nitrogen gun. A 30 nm layer of Cr was deposited by sputtering to form a mask for the following etching step. A subsequent lift-off step with acetone produced the Cr etch mask patterned with nanoarcs. Afterward, reactive-ion etching (Oxford Instruments, PLASMALAB100 ICP380) using 60 sccm of SF6 was performed to transfer the patterns on the mask to the a-Si layer. The remaining Cr mask was removed in standard Cr etchant. The images of the resulting structures were obtained by scanning electron microscopy (FEI, NovaNanoSEM 430).

**FDTD simulations:** 3D simulations probing the resonance modes and chiral beamsplitting are performed using the FDTD method (FDTD solution package from Lumerical Inc.). In the simulations, we import the measured complex refractive index of the amorphous-silicon film characterized by an ellipsometer into the material database of FDTD solution software. We apply the total-field scattered-field source and the analysis group to calculate the scattering cross-section. As for the chiral beamsplitting simulations, we apply the periodic boundary conditions at the x- and y-boundaries and perfectly matched layers at the z-boundary.
Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Figure 1. (a) Artist's concept of broadband chiral beamsplitting achieved by the continuous nanoarcs. When the broadband spectrum source impinged on the nanoarc array, the components of right- and left-handed polarization (corresponding to the blue and red spirals arrow) undergo the axis-symmetric skewing, which is closely analogous to the spin-Hall effect. (b) Schematic view of a unit cell of dielectric nanoarc structure. The geometric parameters are as follows: $R = 450$ nm, $W = 90$ nm, and $H = 190$ nm. The lattice spacings of the array are: $P_x = 1000$ nm and $P_y = 500$ nm. The $u$ and $v$ represent the tangent and normal directions of nanoarc at the localized area. (c) Geometric phase stemming from the nanoarc with a curved path $\zeta$. (d) Simulated scattering cross-section excited by different polarized lights when the nanoarc degenerates into a nanoline at the infinitesimal location. The insets are magnetic field...
distribution for \( u \)-polarized illumination and electric field distribution for \( v \)-polarized illumination at the wavelength of 800 nm. The \( u \) and \( v \) represent the tangent and normal directions of nanoarc at the localized area.

Figure 2 Numerically calculated (a) transmission coefficient \( T_-\sigma_- + T_-\sigma_+ \) and (b) conversion efficiency \( T_-\sigma_- / (T_-\sigma_- + T_-\sigma_+) \) of a-Si:H nanoarcs for a varying structural height \( H \) at a constant width of 90 nm. Numerically calculated (c) transmission coefficient and (d) conversion efficiency for a varying structural width \( W \) at a constant height of 190 nm. \( \sigma = \pm 1 \) denotes RCP and LCP respectively. Incoming light is RCP light in these simulations.
Figure 3 Chiral beamsplitting of nanoarcs. (a) Top-view scanning electron micrograph (SEM) of the chiral beamsplitting a-Si:H nanoarc array. Scale bar is 1 μm. Phase gradient along the interface \( \frac{d\phi}{dx} \) is introduced by the nanoarc. (b) Phase of the Ex component within a unit cell of the transmitted field where the full gradient phase (0-2π) is accomplished by the continuous nanoarc and \( \theta \) is the angle of transmission. (c) Simulated result of the helicity-dependent deflection in the momentum space at the wavelength of 800 nm. \( \sigma = \pm 1 \) denotes RCP and LCP lights. The inserts reveal the far-field intensity distribution. (d) Simulated and experimental results of absolute efficiency and conversion efficiency over a broadband spectrum. (e) Experimentally measured helicity-dependent deflection in the momentum space at the wavelength of 800 nm.
Figure 4 Chiral-detour broadband phase holograms. (a) Schematic illustrating the wavefront modulation of chiral-detour phase principle. The array is composed of numerous a-Si:H-nanoarcs on a glass substrate. Each nanoarc at different locations, $\delta_1, \delta_2, \delta_3, \ldots$, introduces a phase shift $\Delta \varphi$. From the view of the diffraction angle, i.e., $\theta = \arcsin(\lambda / P_x)$, the arbitrary wavefront can be realized by adjusting the locations of the nanoarcs. (b) Target image of a
'Chinese knot' pattern. (c) Phase distribution designed to generate the target holographic image in the far field. (d) SEM image of the nanoarc hologram. Scale bar is 3 μm. (e)-(h) Far-field experimental images generated by the nanoarc metasurface at the wavelengths of 600 nm, 700 nm, 800 nm, and 900 nm, respectively, showing its broadband functionality.

Figure 5 Helicity-dependent holograms. (a) SEM image of two symbols-encoded holograms. The nanoarcs are rationally designed in two parts: in blue, phase information is imparted to those structures to generate the image illustrating the spin state of $\sigma_+$ and phase information is imparted to those structures in red to generate the image illustrating the spin state of $\sigma_-$. The images on the right are numerically reconstructed from the target patterns. (b) Experimental images are generated from helicity-dependent nanoarc at the wavelength of 800 nm. The pattern of $\sigma_-$ shows up when the LCP light impinges on the nanoarc and the pattern of $\sigma_+$ shows up when the LCP light impinges on the nanoarc. Linearly polarized light produces two patterns simultaneously.
Nanoarc metasurface holds promise for simultaneous broadband and high efficiency applications. A few compelling functionalities, such as chiral beamsplitting, broadband holography, and helicity-selective holography, can be achieved by dielectric nanoarcs. The continuous nanoarc metasurfaces prevail over plasmonic or dielectric discretized building block strategies and they can lead to novel designs of spin-controllable metadevices.
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