

Synthesis of broadband multilayer metamaterial absorbers based on spatially variable 3D-printed structures and MXenes

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We introduce a systematic approach for enhancing the absorbance of multilayer metamaterial absorbers (MMA) by combining deep-subwavelength spatially variable dielectric substrates with the increased Ohmic losses of MXene resonators. We choose MXenes to form the MMA resonators as they are ultra-thin conductive materials that offer higher Ohmic losses than copper used in PCBs, alleviate the need for the post-treatment required by metal inks, and are considerably thinner than conductive filaments. Previous multilayer MMAs in the literature typically involve structured metal layers with a uniform dielectric profile or pyramidal-shaped substrates. The simplicity of the dielectric structure limits the design freedom and operational bandwidth of MMAs. In this work, we explore an alternative route to achieving increased design flexibility and enhanced absorption, by tailoring the dielectric substrates and consequently the underlying physics on a deep subwavelength scale. Advances in stereolithography (SLA) three-dimensional (3D) printing, allow the synthesis of suitable spatially variable 3D-printable dielectric structures. We inversely design the multilayer MMA by topology optimization, with the MMA substrate cells assuming dielectric constant values in the continuous range between air and the 3D printer's resin. We develop a simple postprocessing methodology, BINACONN3D, that transforms the optimized non-manufacturable MMA into a manufacturable one, while preserving performance. The BINACONN3D uses manufacturable configurations of a prescribed local air and resin composition to realize each designed dielectric material and accommodates detailed connectivity constraints across consecutive layers, to allow multilayer fabrication and assembly. Our work demonstrates the advantages of combining inversely designed spatially variable dielectric structures with the increased Ohmic losses of MXene resonators. We fabricate and experimentally characterize the synthesized MMA, with measurements in good agreement with the simulations. Our approach paves the way for improving the performance of existing metamaterial devices by leveraging high-resolution spatially variable dielectric structures.

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I. INTRODUCTION

In recent years, metamaterials, which are artificial materials that can control electromagnetic waves, have gained increasing interest in numerous applications, such as absorbers, gratings, and antennas. Landy *et al.* [1] demonstrated a metasurface absorber, which achieves near-unity absorbance close to resonance. Broadband absorption is essential in many microwave applications, such as solar energy harvesting, radar cross-section

(RCS) reduction, and electromagnetic interference shielding. Various approaches have been used to achieve broadband absorbance by constructing metastructures. Such approaches include superimposing multiple resonant modes with different resonance frequencies to realize the broadband response, a dispersion compensation approach based on metal-dielectric materials, nonresonant effects of the metastructures, lowering the Q factor, and introducing ultrafast systems. [2] Nonuniform multielement metamaterial absorbers have been introduced to superimpose multiple resonant modes with different resonance frequencies. [3] The resonators can be located on the same plane [4] or in different layers, forming a multilayer absorber [5–7].

Multilayer MMAs are traditionally based on printed circuit boards (PCBs) [5,6], which limits the absorbance due to the relatively low Ohmic losses of copper, and has a complex stacking process. Conductive materials with increased Ohmic losses, such as conductive inks

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[8], filaments [9], or indium tin oxide (ITO) [10], can offer enhanced absorption compared to PCBs. However, metal inks require post-treatment, whereas conductive filaments are relatively thick, on the order of 0.3 mm, and may also lead to precision degradation when combined with dielectric materials of different glass temperatures [9]. Enhancing the absorption of MMAs and moving toward Rozanov's limit is an ongoing effort that will benefit RCS reduction applications. To further enhance the absorbance of MMAs, optimized complex shapes have been synthesized on the metal surfaces [11]. However, tailoring the properties of the MMA's dielectric substrates to form spatially variable dielectric profiles of complex optimized shapes has not yet been explored. Tailoring the underlying physics of the dielectric structures on a deep sub-wavelength scale offers an alternative route to achieving increased design flexibility and enhanced absorption.

Transition metal carbides (TMCs), also known as *MXenes*, are two-dimensional conductive materials, which present an alternative to metal inks and 3D-printed conductive filaments [12]. *MXenes* alleviate the need for additives and post-treatment of metal inks, are considerably thinner than conductive filaments, and offer lower conductivity and increased Ohmic losses compared to copper-based alternatives. Recently, *MXenes* have been employed to realize MMAs, with applications ranging from microwave to infrared frequencies. [13–15]

Advances in 3D-printing technology have enabled the realization of various easily manufacturable devices [16, 17]. Filament-based 3D printing has been used primarily for the fabrication of MMAs, such as single-layer devices, with a resistive ink deposited on a 3D-printed perforated substrate, to achieve a lightweight design [8]. Multilayer 3D-printed MMAs have also been realized, with interleaved conductive and dielectric filament layers to form the multilayer stack [9]. Although 3D printing has been used in designing certain MMAs, applications are limited to substrates with uniform dielectric properties. Stereolithography (SLA) 3D printing presents a higher precision alternative to filament-based 3D printing, and has mainly been used in manufacturing all-dielectric lenses [18,19]. The ability of manufacturing high-precision dielectric structures has not yet been harnessed to enhance MMA absorption.

To enhance the absorbance of MMAs, topology optimization approaches can be employed. In multilayer MMAs, both the metal surfaces and the dielectric substrates can benefit from such approaches. Topology optimization has been used to tailor the properties of metal surfaces, usually in conjunction with genetic algorithms [11,20,21]. Inverse design approaches, such as objective first [22] and adjoint optimization [23], have been used to synthesize devices with enhanced efficiency. Such methodologies have been applied to all-dielectric devices and are based on gradient-based approaches. Inverse design

methodologies usually result in complex-shaped dielectric profiles, with the dielectric constant varying in a continuous range. Binarization and connectivity constraints can be added to the optimization, in the form of binarization cost, artificial attenuation, or filtering constraint [23]. A simple postprocessing methodology, the BINACONN methodology, was introduced as an alternative [24]. In that case, the topology optimization does not include fabrication constraints, and is therefore simpler and may also lead to an optimized device with higher performance. The BINACONN methodology transforms the optimized device of a continuous dielectric profile into a manufacturable one, while preserving the device's performance as close as possible to the continuous case. Although this methodology presents a simple approach to rendering optimized devices manufacturable, it has only been demonstrated on two-dimensional devices with basic connectivity constraints. As the complexity of devices increases, especially in the context of three-dimensional multilayer devices, fabrication constraints also become more complex, as they have to be aligned with the fabrication and assembly of multilayered complex-shaped structures.

In this work, we provide a systematic synthesis approach for realizing multilayer MMAs that combine lossy *MXene* resonators and high-resolution spatially variable dielectric layers to enhance the absorption of MMAs with a uniform dielectric profile. Our synthesis approach involves two interleaved steps, which can be performed iteratively; tailoring the *MXene* surfaces and the dielectric layers. As a proof of concept, we consider simpler *MXene* patch resonators on each layer of the multilayer MMA and focus on tailoring the properties of the dielectric layers to illustrate the advantages of spatially variable dielectric substrates. Our primary goal is to demonstrate the synthesis approach of spatially variable dielectric structures, rather than a specific engineering design. Inverse design is employed to engineer the underlying physics of the supported resonances and achieve devices of enhanced absorbance. We perform topology optimization [23] in all dielectric layers, where each substrate cell can assume continuous values between the resin and the air. We introduce a postprocessing methodology, BINACONN3D, to transform the optimized spatially variable MMA into a manufacturable one, while preserving performance. The BINACONN3D accommodates suitable complex fabrication constraints across consecutive layers that facilitate the assembly and fabrication of multilayer MMAs. The optimized manufacturable MMA is fabricated and experimentally characterized in the X-band, to provide additional justification.

II. METHODOLOGY OVERVIEW

We provide an overview of the methodology followed to synthesize manufacturable MMAs with a spatially variable material profile. Figure 1(a) shows an MMA with *MXene*

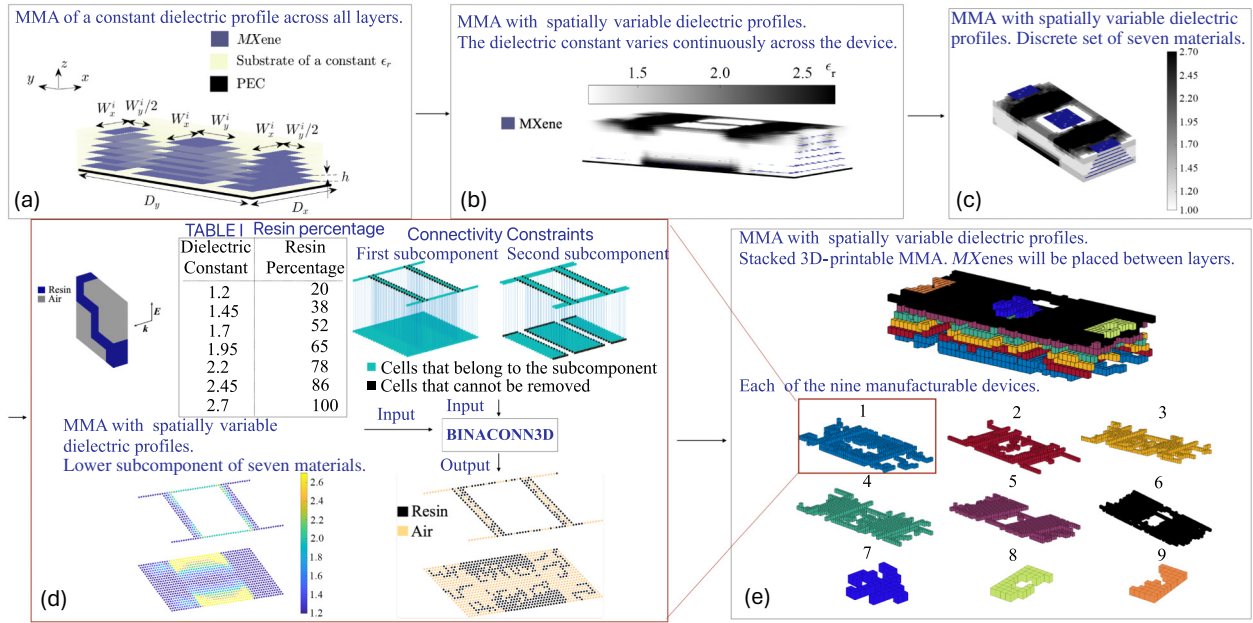


FIG. 1. Methodology overview. (a) Multilayer MMA of a constant dielectric profile. (b) MMA with a spatially variable dielectric profile. The dielectric constant varies continuously across each layer. This design is obtained by TopOpt. (c) The MMA of (b) is transformed into an MMA of a discrete set of materials, i.e., seven materials and remains nonmanufacturable. (d) BINAConn3D takes each subcomponent of (c) and transforms it into a manufacturable one, taking into account the resin percentage of each material and connectivity constraints. (e) Each of the nine manufacturable subcomponents is shown both as stand alone and stacked.

patches and a uniform dielectric constant profile across all layers, which serves as a baseline for the MXene MMA with a spatially variable material profile. Our synthesis process involves the following steps:

(1) We synthesize an MMA with a spatially variable material profile by inverse design, using density-based topology optimization. The MXene patches remain the same as in the baseline MMA; we vary the material properties of the dielectric layers. We assume two available materials, Formlabs 3D-printer's Clear V4 resin, and air ($\epsilon_r = 1$). In Ref. [25], the Clear resin is measured with a relative permittivity of $\epsilon_r = 2.7$ obtained at 12 GHz and $\epsilon_r = 2.65$ at 20 GHz. The loss tangent is measured at 0.03 at both frequencies. As the variation between frequencies is small, we adopted $\epsilon_r = 2.7$ across all frequencies $f = [8, 25]$ GHz for simplicity. We have also included the simulated absorbance of the manufacturable optimized MMA with $\epsilon_r = 2.65$ adopted above 16 GHz in Sec. III of the Supplemental Material [26]; the influence of changing ϵ_r is almost negligible. The dielectric layers consist of subwavelength cells of size about $1/40$ of the free space wavelength at the center frequency. The optimized device is obtained by allowing each cell's relative dielectric constant to continuously vary within the range of available dielectric materials and is shown in Fig. 1(b).

(2) Our next steps are to transform the nonmanufacturable MMA with a spatially variable dielectric constant

profile into a manufacturable one, while preserving performance; therefore, we develop the BINAConn3D methodology. The steps followed to implement BINAConn3D include the following preprocessing steps:

(a) Transforming the optimized MMA with a continuous spatially variable dielectric profile into a discrete one, by keeping as many material levels needed to preserve high absorbance. The device of a discrete number of materials remains nonmanufacturable [Fig. 1(c)].

(b) Assigning a resin percentage to each intermediate material level, as shown in Fig. 1(d)'s Table I. We assign a resin percentage instead of a single configuration of predetermined shape to each material level, to enable greater design flexibility, which is useful for devices with conformal and complex-shaped dielectric materials, as the one in Fig. 1(c). Resin percentage does not exclusively define the effective dielectric constant. Structures of a uniform or close to uniform air and resin distribution have an effective propagation constant very close to the average of all air and resin distributions [24]. Unit cells with a uniform air and resin distribution, i.e., finer internal structures, are more suitable to approximate the assigned dielectric constant. Hence, we consider close-to-uniform configurations that are also manufacturable, as is the unit cell shown in Fig. 1(d), to determine the suitable resin percentage. For each intermediate material level, we select the resin percentage for which the propagation constant of the mode supported by the air and resin configuration matches that of the intermediate material level.

(c) Identifying suitable connectivity constraints to assemble the multilayer MMA. As shown in Fig. 1(d), the multilayer MMA will be assembled by selectively extruding regions of each lower dielectric layer and forming complementary gaps in the adjacent upper layer to ensure precise vertical alignment and structural integration. Additional details on the fabrication constraints will be provided in the following sections.

(3) The BINACONN3D is applied to each subcomponent of the multilayer MMA. Each subcomponent must remain intrinsically connected. The BINACONN3D substitutes nonmanufacturable materials with manufacturable structures of the suitable resin percentage that comply with the fabrication constraints. The output of BINACONN3D is a manufacturable subcomponent, as the one shown in Fig. 1(d).

(4) The assembly of the manufacturable subcomponents in a multilayer MMA is shown in Fig. 1(e).

III. INITIAL MMA WITH A CONSTANT DIELECTRIC PROFILE

A. Single-layer MMA

We consider a baseline multilayer metal-backed MMA with *MXene* patches as the resonators and a uniform dielectric profile across all layers, which serves as a point of comparison for the MMA with spatially variable dielectric structures. Since we design the MMA to fit within and be measured in a WR90 waveguide, the simulations of the single-layer and multilayer MMA of a constant dielectric material, as well as the optimization and BINACONN3D simulations of Secs. IV and V, are all performed using the TE₁₀ mode as excitation with perfect electric conductor boundary conditions (PECs) on the side boundaries. Hence, our MMA will be optimized only for the TE₁₀ mode excitation of the WR90 without considering any polar-angle dependence. In Sec. I of the Supplemental Material [26], we demonstrate that the synthesis process of spatially variable dielectric substrates and the BINACONN3D can also be extended to design optimized polarization-insensitive MMAs that have equal absorbance for *x*- and *y*-polarized normally incident plane waves. As shown in Fig. 2, each MMA layer consists of three rectangular patches. We tailor the rectangular patch dimensions of a single-layer MMA to obtain resonances that cover the X- and Ku-bands. Individual single-layer MMAs will be assembled to synthesize the multilayer MMA. Each *MXene* rectangular patch behaves as an *LC* resonator; we can tune its resonance frequency by varying the patch dimensions or the substrate properties. The thickness of each single-layer MMA is $h = 0.6$ mm and the substrate's dielectric constant is $\epsilon_r = 2.7(1 - j0.03)$. In this initial design phase, we also set the *MXene* patches as PEC and focus only on the resonance frequency. The dimensions of the center patch are $W_x = Np_x$ and $W_y = Np_y$ along the

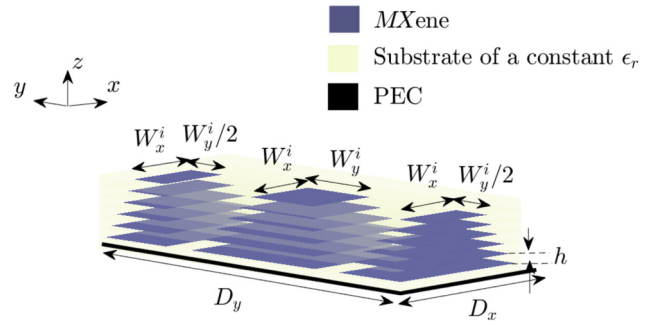


FIG. 2. Multilayer MMA comprised of *MXene* patches and dielectric substrates of a constant material profile.

x and *y* axes, where N is an integer, $p_x = D_x/22$, $p_y = D_y/48$, and $D_x = 10.16$ mm, $D_y = 22.86$ mm. The off-center patches have the same length W_x as the center patch along the *x* axis and half the width W_y along the *y* axis. The simulation setup described in the Appendix is adopted for the FEM simulations. The simulated absorbance of a single-layer MMA is provided in Fig. 3, varying the length and width of the patches as a function of N . By varying N between 10 and 20 we obtain resonances between 8.5 and 16.5 GHz. Figure 3 shows the absorbance both considering only the center patch as well as with the addition of the off-center patches. As the MMA is excited by a TE₁₀ mode, the center patch exhibits higher absorbance, with the off-center patches exhibiting a lower absorbance level.

B. Multilayer MMA of a uniform material profile

We combine individual single-layer MMAs to form a multilayer MMA. We consider six *MXene* layers with the *MXene* patches forming pyramids, i.e., the largest patches are arranged on the lower layer and the smallest patches on the top layer, and seven dielectric layers, with the seventh layer covering the top *MXene* patches. The seventh dielectric layer is added to facilitate manufacturing,

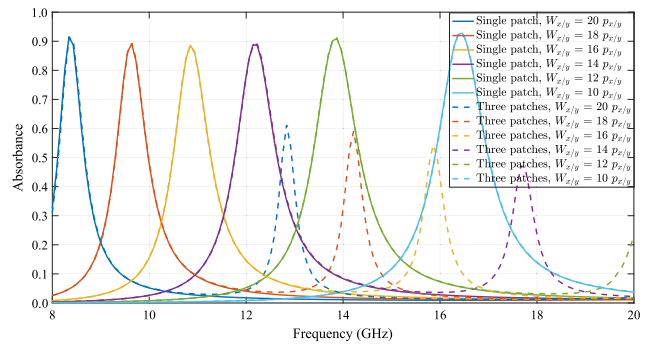


FIG. 3. Simulated absorbance for a single-layer MMA with one center (solid lines) and three, center and off-center (dashed lines) *MXene* patches, varying the patch width $W_{x/y}$.

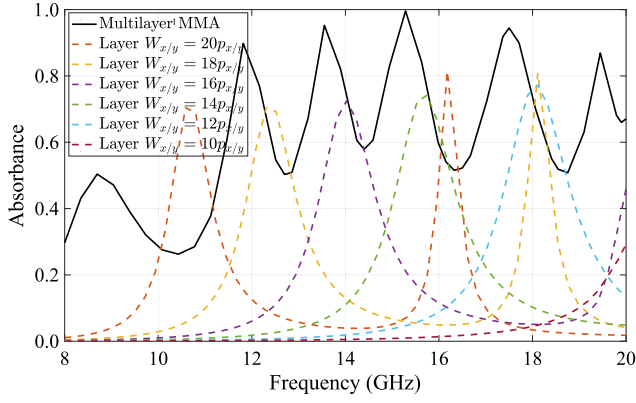


FIG. 4. Simulated absorbance for a multilayer MMA with a uniform substrate of $\epsilon_r = 1.5(1 - j0.03)$ and PEC on the MXenes vs. the absorbance of each layer individually.

as will be shown in the following sections. We demonstrate that layer-layer interaction influences the multilayer MMA's absorbance. We consider a dielectric constant of $\epsilon_r = 1.5(1 - j0.03)$ across all layers and impose PEC boundary conditions on all patches. In Fig. 4, we observe that the response of the multilayer MMA (black, solid line) cannot be reconstructed from the individual responses of each MMA layer (dashed, color lines), since the resonance frequencies and the strength of each resonance vary considerably. The multilayer MMA has a higher absorbance at each resonance frequency than each layer independently. This shows that mutual coupling between layers significantly contributes to the multilayer MMA response.

1. Assessing the effect of varying the MXene conductivity

The electrical conductivity of the MXenes can be modified by a plasma-assisted atomic layer etching (ALE) process [12]. This process can modify the MXene's surface chemistry, converting the surface terminations from fluorine dominated to oxygen dominated. Plasma treatment increases the conductivity of MXene films, as plasma ALE can effectively remove conductivity-reducing terminations (e.g., $-F$). Figure 5(a) shows an MXene sample deposited on a polycarbonate membrane and Fig. 5(b) depicts the atomic structure model of Ti_3C_2Tx MXene, showing titanium (Ti), carbon (C), and surface terminations (Tx) such as $-OH$, $-O$, and $-F$.

We measured both plasma-treated and non-plasma-treated MXene samples in the X-band, and estimated the surface resistance between $R_s = 7\Omega/\text{sq}$ and $R_s = 12\Omega/\text{sq}$, corresponding to bulk conductivity values between $9.26 \times 10^4 \text{ S/m}$ and $1.59 \times 10^5 \text{ S/m}$. We confirm that plasma-treated MXene samples have a higher bulk conductivity than non plasma-treated ones in the X-band. The MXene bulk conductivity is considerably lower than the copper conductivity, which is about $5.8 \times 10^7 \text{ S/m}$, offering the potential for synthesizing MMAs

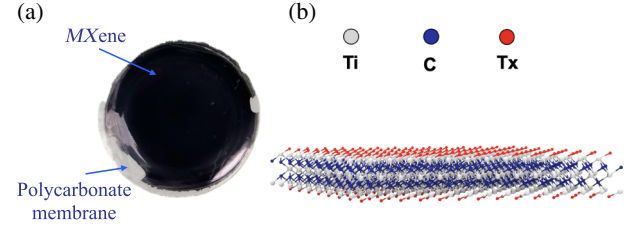


FIG. 5. (a) An MXene sample deposited on a polycarbonate membrane. (b) Atomic structure model of Ti_3C_2Tx MXene, showing titanium (Ti), carbon (C), and surface terminations (Tx) such as $-OH$, $-O$, and $-F$.

with increased Ohmic losses compared to PCB alternatives. We note that the surface resistance was measured exclusively in the X-band. The X-band surface resistance value is also used for simulations at higher frequencies up to 25 GHz. The actual Ku-band surface resistance value may deviate from the one used in the simulations; however, this assumption was necessary due to equipment restrictions. Setting different MXene conductivity values in the Ku-band would not impact the proposed design process, as they can be incorporated into the simulations.

Each multilayer MMA patch can have a different surface resistance value, which provides additional design freedom. At this point, we set all MXene patches to have the same surface resistance value and vary R_s between $7\Omega/\text{sq}$ and $12\Omega/\text{sq}$. The absorbance for frequencies between 8 and 20 GHz, covering and slightly extending over the X- and Ku-bands, is shown in Fig. 6. As the range of R_s values is relatively narrow, the effect of changing the surface resistance is negligible. We select $R_s = 12\Omega/\text{sq}$, which achieves slightly higher absorbance in the X-band. We also compare the absorbance of the MXene-based MMA with that of the corresponding MMA, where PEC boundary conditions are imposed on all metal surfaces. We verify the benefit of using lossy MXenes instead of lossless metals, since MXenes provide a wide absorption bandwidth in comparison to the narrow peaks provided by the PEC resonators.

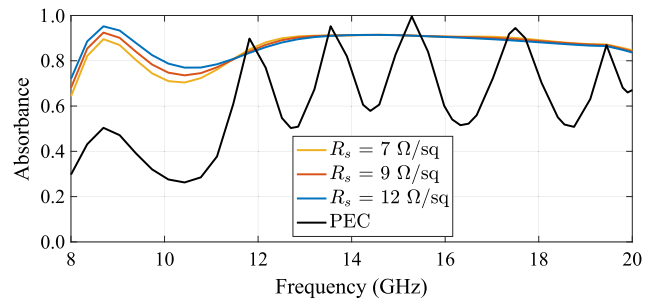


FIG. 6. Simulated absorbance for a multilayer MMA with a uniform substrate of $\epsilon_r = 1.5(1 - j0.03)$, varying the MXene's surface resistance R_s .

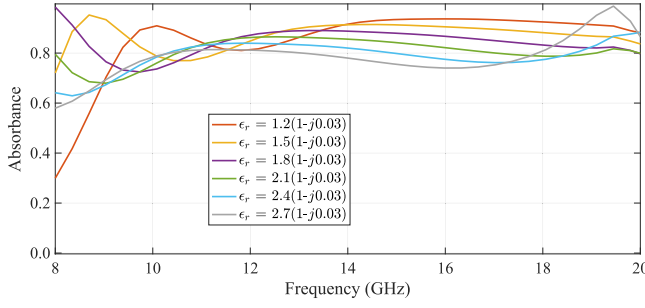


FIG. 7. Simulated absorbance for a multilayer MMA varying the substrate relative dielectric constant ϵ_r for $R_s = 12 \Omega/\text{sq}$.

2. Assessing the effect of varying the effective dielectric constant

The effective dielectric constant of the 3D-printed dielectric substrates can also be modified by engineering suitable subwavelength structures that achieve the desired effective property [27]. Each subwavelength structure is composed of air and the 3D printer's resin. We assume uniform dielectric properties across all layers and vary the dielectric constant, with the real part assuming values within [1.2, 2.7]. In Fig. 7 we observe that higher effective dielectric constant values shift the resonances to higher frequencies. The effective dielectric constant of $\epsilon_r = 1.5(1 - j0.03)$ leads to the highest average absorbance between 8 and 20 GHz. We show the normalized magnetic-field intensity across $f = \{9, 11, 13, 15, 17, 19\}$ GHz in Fig. 8 and observe that the modes supported by the multilayer MMA extend over multiple layers, due to the resonator dimensions varying slowly between neighboring layers [6]. This verifies the presence of layer-layer interaction. Since the modes extend over multiple layers, modifying the properties of the dielectric substrate has the potential to improve the absorbance across a wide frequency range. The uniform multilayer MMA considered in this section serves as a baseline example, which will be used to demonstrate the advantages of using spatially variable dielectric structures over uniform ones in Sec. IV. Considering a

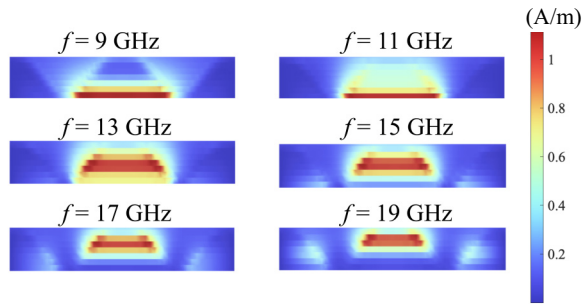


FIG. 8. Normalized magnetic-field intensity (A/m) across various frequencies for a multilayer MMA of $\epsilon_r = 1.5(1 - j0.03)$ across all layers and $R_s = 12 \Omega/\text{sq}$.

greater total thickness can increase absorption, as dictated by Rozanov's limit [28], as does adding additional layers in a prescribed thickness [6].

IV. OPTIMIZED MMA WITH SPATIALLY VARIABLE DIELECTRIC SUBSTRATES

We inversely design the MMA by performing a density-based topology optimization with the TopOpt method [23]. The dielectric constant of all substrates can continuously vary in a specified range. The TopOpt method uses adjoint sensitivity analysis to perform gradient computations, which requires only solving a single (adjoint) equation for the optimization objective and one for each constraint in the optimization problem, independent of the size of the design space. We aim to maximize absorbance A . The optimization problem is formulated as follows:

$$\begin{aligned} \max_{\xi(\mathbf{r})} & A(\mathbf{E}(\mathbf{r}_p), \varepsilon(\xi(\mathbf{r}))) \\ \text{s.t. } & \mathcal{L}_{\text{EM}}(\mathbf{E}(\mathbf{r}), \varepsilon(\xi(\mathbf{r}))) = \mathbf{E}_{\text{in}}(\mathbf{r}_p), \\ & \varepsilon(\xi(\mathbf{r})) = \varepsilon_{r,\text{res}} + \xi(\mathbf{r})(\varepsilon_{r,\text{air}} - \varepsilon_{r,\text{res}}), \\ & 0 \leq \xi(\mathbf{r}) \leq 0.8824. \end{aligned} \quad (1)$$

The operator $\mathcal{L}_{\text{EM}}$ expresses the wave equation in terms of the electric field and $\mathbf{E}_{\text{in}}(\mathbf{r}_p)$ is the incident electric field on the port plane.

$$\mathcal{L}_{\text{EM}}(\mathbf{E}(\mathbf{r}), \varepsilon(\xi(\mathbf{r}))) = \nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - \frac{\omega^2}{c^2} \varepsilon_r(\mathbf{r}) \mathbf{E}(\mathbf{r}). \quad (2)$$

We choose to maximize absorption A over a broad frequency range from 8 to 25 GHz, hence we consider the sum of the objectives, i.e., the absorption, over 25 considered frequencies. The absorbance A is expressed in terms of the S parameters S_{i1} , as $A = 1 - \sum_{i=1}^N |S_{i1}|^2$, where S_{11} is the reflection coefficient, and S_{i1} , with $i > 1$, are the S parameters of the higher-order waveguide modes. Additional information on the calculation of absorption in the FEM simulations is available in the Appendix. The continuous design field $\xi(\mathbf{r})$ is used to interpolate the dielectric constant between $\epsilon_r = 1.2$ ($\xi(\mathbf{r}) = 0.8824$) and the 3D printer's resin $\epsilon_r = 2.7$ ($\xi(\mathbf{r}) = 0$). We limit the range from [1, 2.7] to [1.2, 2.7], to avoid disconnected components.

The dielectric constant profile of the optimized spatially variable MMA is shown in Fig. 9 both in a three-dimensional view and in the center of each dielectric layer. We observe that the dielectric forms complex, conformal, and nonintuitive shapes and assumes continuous values in the range [1.2, 2.7]. We compare the absorbance of the spatially variable MMA to the MMAs with uniform substrate properties. The average absorbance of the spatially variable MMA is $A_{\text{avg}} = 94.07\%$ between 8 and 25 GHz, which is considerably higher than that of all MMAs with

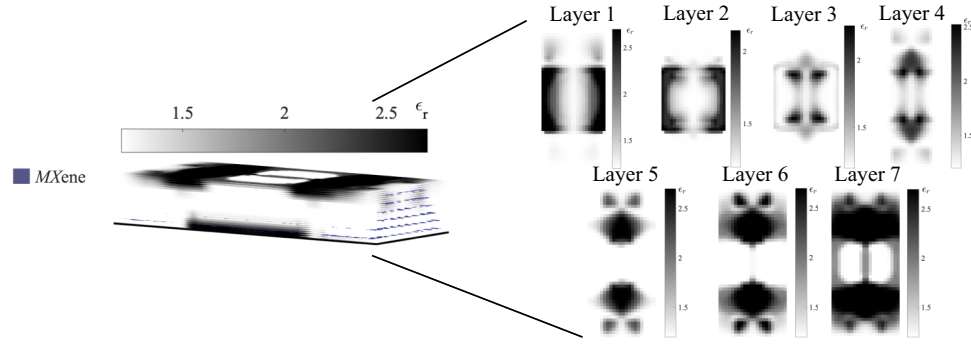


FIG. 9. The spatially variable dielectric constant profile of the optimized MMA at each dielectric layer. The dielectric constant has dielectric constant values in a continuous range between $\epsilon_r = 1.2(1 - j0.03)$ and $\epsilon_r = 2.7(1 - j0.03)$.

uniform substrate properties, the highest of which is $A_{\text{avg}} = 83.82\%$ in the same band.

Our synthesis approach is based on inverse design; therefore, we do not directly get a detailed physical explanation of why the optimized MMA with spatially variable dielectric layers achieves higher absorbance than the MMA with constant dielectrics. Instead, we attempt to gain insight into the broadband absorbance mechanisms by observing the simulated fields of the optimized and nonoptimized MMA. Perfect absorption at a certain resonance frequency is achieved when the critical coupling condition is fulfilled, i.e., the external decay rate is equal to the intrinsic decay rate [29]. In Sec. IV of the Supplemental Material [26], we calculate the intrinsic and external decay rates at the resonance frequencies of an optimized and nonoptimized symmetric MMA, for modes excited by a normally incident plane wave, and check which MMA is closer to fulfilling the critical coupling condition. We observe that the resonances of the optimized spatially variable MMA are more uniformly spaced within the 8–25 GHz range than those of the MMA of a constant material. The resonances of the optimized MMA have intrinsic and external decay rates closer to each other, i.e., closer to satisfying the critical coupling condition. The insight gained by the above observations can be used as the basis for the considered

nonsymmetric optimized spatially variable MMA, excited by the TE_{10} waveguide mode.

The normalized magnetic-field intensity is shown across various frequencies for the spatially variable MMA in Fig. 11. We observe that the modes supported by the optimized spatially variable multilayer MMA also extend over multiple layers, and are more smoothly distributed across all layers, compared to the MMA of a uniform dielectric constant. As a next step, we will transform the spatially variable dielectric substrates of a continuous profile into a discrete one, keeping the necessary number of material levels.

V. TRANSFORMING THE MULTILAYER MMA OF A SPATIALLY VARIABLE DIELECTRIC PROFILE INTO A MANUFACTURABLE ONE

A. MMA with a discrete number of materials

A first step toward transforming the nonmanufacturable multilayer MMA of a spatially variable dielectric profile into a manufacturable one is to preserve a discrete number of dielectric constant values. We examine the cases of preserving two, four, and seven material levels. Each dielectric constant value is substituted by the closest discrete value. In Fig. 12 we compare the absorbance to the

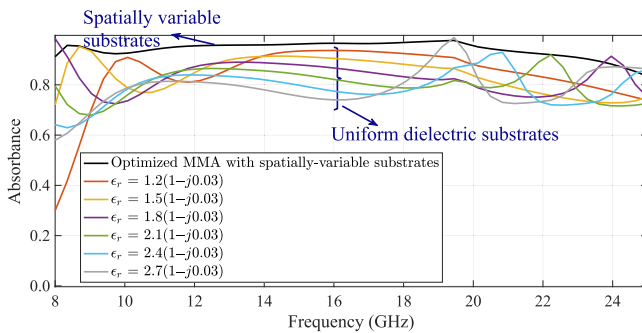


FIG. 10. Simulated absorbance comparison of the MMA with spatially variable and uniform dielectric layers.

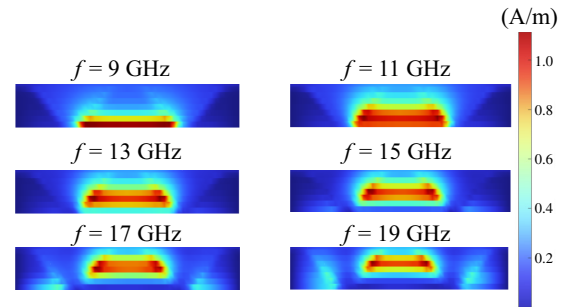


FIG. 11. Normalized magnetic-field intensity (A/m) across various frequencies for the optimized device of a continuous dielectric profile.

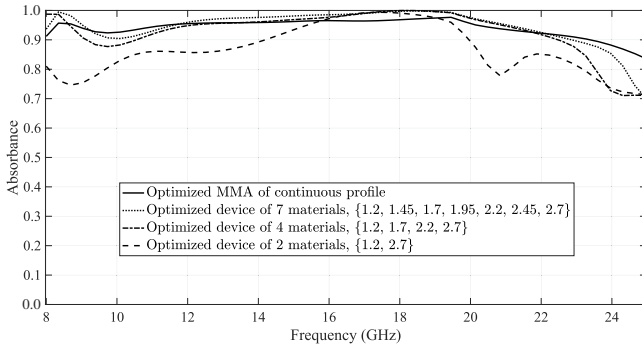


FIG. 12. Comparison of the simulated absorbance of both the optimized MMA with the dielectric constant having a continuous range, as well as two, four, and seven materials.

MMA obtained by the TopOpt method. We observe that two material levels are insufficient, leading to a reduced absorbance. Seven materials lead to higher absorbance than four. A greater number of materials is not necessary, as the discrete device of seven material levels $\{1.2, 1.45, 1.7, 1.95, 2.2, 2.45, 2.7\}$ has a very close average absorbance to that of the continuous profile, about $A_{\text{avg}} = 94\%$.

B. BINACONN3D

We describe the steps followed to implement the BINACONN3D methodology and especially elaborate on the necessary fabrication constraints that should be imposed to ensure a manufacturable multilayer MMA.

1. Fabrication constraints

We identify suitable subcomponents that must remain connected, to facilitate the multilayer MMA construction. Synthesizing the multilayer MMA as a single connected component is not an option, as the *MXene* resonators must be inserted between each pair of consecutive dielectric layers. To that end, a subcomponent is synthesized by selectively extruding regions of each lower dielectric layer and forming complementary gaps in the adjacent upper layer to ensure precise vertical alignment and structural integration. The extruded regions are defined to accommodate the insertion and precise positioning of the *MXene* patches at designated locations between adjacent dielectric layers. Specifically, the subcomponents are defined as follows:

(a) Subcomponent 1 (adjacent to the metal ground) includes the entire first dielectric layer and the parts of the second dielectric layer that will be extruded and form a frame.

(b) Subcomponents 2–6 include the nonextruded parts of the lower dielectric layer and the extruded parts (frame) of the dielectric layer directly above.

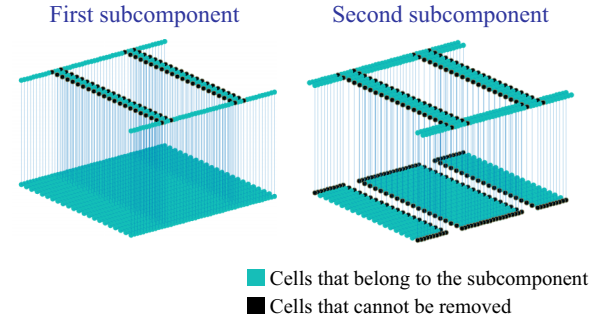


FIG. 13. Graph of subcomponents 1 and 2. Each cell that belongs to the subcomponent is shown as a node (turquoise). Nodes that are immediate neighbors are connected by an edge. The black nodes will remain as resin.

(c) Subcomponents 7–9 contain only the nonextruded parts of dielectric layer 7.

We identify the cells that belong to each subcomponent. Each cell can be represented as a graph node V . Two graph nodes are connected by an edge E if they are immediate neighbors (north, west, south, east). The diagonally placed nodes are not connected. For the seven materials nonmanufacturable MMA, we form a graph $G(V, E)$ of all cells that belong to a subcomponent. The graphs of subcomponents 1 and 2 are shown in Fig. 13. Each cell that belongs to the subcomponent is shown as a node (turquoise). The nodes that are immediate neighbors are connected by an edge in Fig. 13. Our goal is to transform the nonmanufacturable subcomponents to manufacturable ones, preserving the connectivity of each subcomponent. To promote structural integrity, we specify certain cells that must remain as resin, i.e., the black nodes in Fig. 13. This will facilitate the assembly process of consecutive dielectric layers.

2. BINACONN3D algorithm

We implement the BINACONN3D algorithm for each subcomponent, with connectivity constraints imposed on a subcomponent level (Fig. 13). The BINACONN3D assigns air or resin to each cell of $\epsilon_{r,d}$ dielectric constant, according to the prescribed air and resin percentage. The steps of the methodology are as follows:

(a) For each material level $\epsilon_{r,d}$, we form the corresponding graph G_d and locate its graph components, i.e., material regions that are intrinsically connected.

(b) For each graph component i of the material $\epsilon_{r,d}$, we generate manufacturable air and resin configurations.

(c) To generate each manufacturable air and resin configuration, we set n_a cells to air, according to the assigned air and resin percentage.

(i) A uniform random distribution is used to determine the cells that will be set to air in each graph component. We

want air and resin to be distributed more uniformly, i.e., to have configurations with finer features.

(ii) Before setting a cell to air, we check that the MMA subcomponent remains connected upon removal of the cell. If it becomes disconnected, we return to the previous configuration. We also check that the cell does not belong to the set of cells that must remain as resin (black nodes of Fig. 13).

(d) We implement the previous steps on all materials for each MMA subcomponent. We produce multiple manufacturable subcomponents, each with a different random seed.

(e) We select a set of manufacturable subcomponents with an absorbance as close as possible to the nonmanufacturable case.

We note that the BINACONN3D methodology is scalable to even larger scale and more complex devices. In this paper, we simulate the entire manufacturable MMA to select the better performing manufacturable subcomponents. However, for even larger-scale devices, where simulating the entire device can become even more computationally demanding, we can simulate smaller areas of the device instead. We would then select the smaller manufacturable structures that are a better match to their nonmanufacturable counterparts. We would then combine all smaller manufacturable structures together to attain the entire manufacturable device, with the performance simply verified by measurements. A proof of concept of this localized approach is briefly discussed in Ref. [30].

C. Assessing the absorbance of the manufacturable subcomponents

We employ the BINACONN3D algorithm and obtain several manufacturable MMAs. A manufacturable MMA is shown in Fig. 14(a), with each subcomponent depicted in a different color. To illustrate that each subcomponent is intrinsically connected, we show each resin cell as a node with immediate neighbors connected by an edge. The connected edges are visible both in-plane and across adjacent layers. We also show each manufacturable subcomponent separately in Fig. 14(b).

We simulate eight manufacturable MMAs and show the absorbance between 8 and 25 GHz in Fig. 15. All manufacturable devices have very similar average absorbance values, between 93.01% and 93.46%. The average absorbance of all manufacturable devices is also very close to the average absorbance of the nonmanufacturable MMA with seven material levels, which is 94.39%. The manufacturable MMAs have absorbance over 90% between 8 and 22 GHz.

We calculate the theoretical minimum MMA thickness $d_{\min}$ required to achieve the absorbance bandwidth of the manufacturable spatially variable MMAs in Fig. 15,

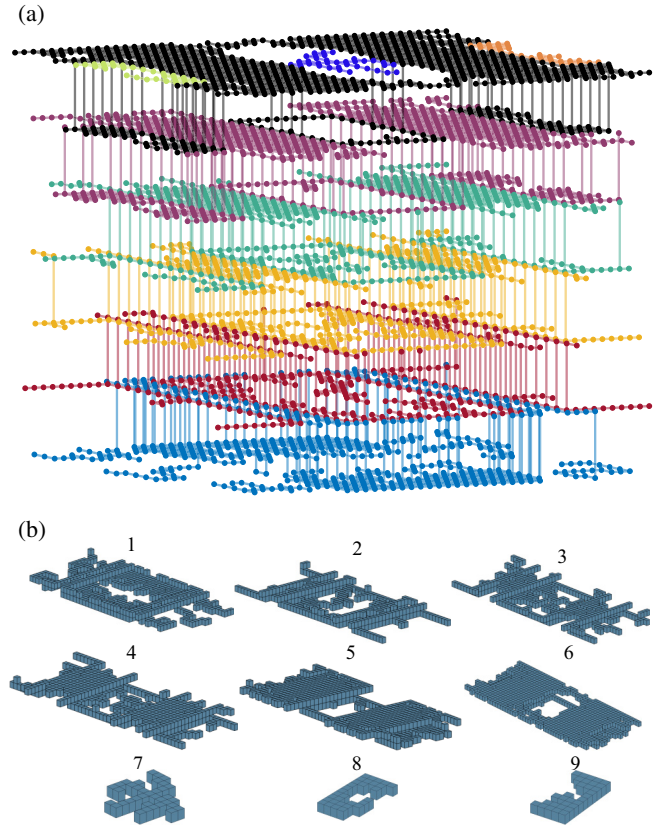


FIG. 14. All nine manufacturable subcomponents. (a) Graph concept. Each resin cell is shown as a node. An edge connects two nodes, only if the nodes are immediate neighbors. Each manufacturable subcomponent is shown in different color. (b) Resin cells are shown in blue color for each subcomponent. Each subcomponent is intrinsically connected.

according to Rozanov's limit [28]. The minimum thickness is calculated by the following inequality as

$$d_{\min} \geq \frac{\left| \int_{\lambda_{\min}}^{\lambda_{\max}} \ln |\Gamma(\lambda)| d\lambda \right|}{2\pi^2}, \quad (3)$$

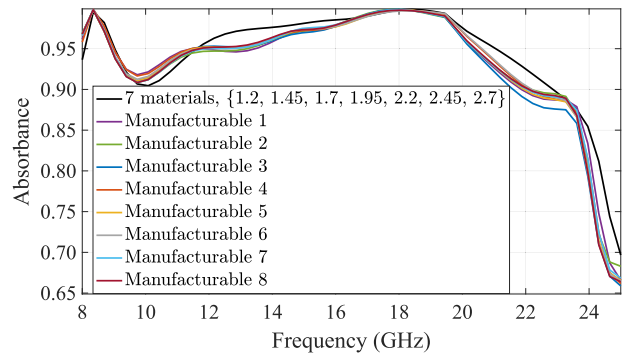


FIG. 15. Simulated absorbance comparison of both the optimized nonmanufacturable MMA with seven materials and the manufacturable MMAs.

for nonmagnetic materials, where $\Gamma(\lambda)$ is the reflection coefficient corresponding to MMAs of Fig. 15 within the operating waveband. The operating waveband is defined by $\lambda_{\min}$ at 25 GHz and $\lambda_{\max}$ at 8 GHz. The calculated minimum theoretical thickness is $d_{\min} = 2.2$ mm, which is close to two times less than the thickness of the MMAs ($h_{\text{tot}} = 4.2$ mm). The quantity $\eta = h_{\text{tot}}/d_{\min}$ quantifies how close the MMA thickness h_{tot} is to the minimum thickness $d_{\min}$ dictated by Rozanov's limit. For the synthesized manufacturable, spatially variable MMA η is $\eta_{\text{s.v.}} = 1.909$, while for the uniform MMA of Fig. 10 with $\epsilon_r = 1.5(1 - j0.03)$ it is $\eta_{\text{uni}} = 3.23$. We conclude that MMAs with spatially variable dielectric layers have lower η values than those with uniform dielectrics; hence, the introduction of variable dielectric layers helps us to move closer to Rozanov's theoretical limit. Approaching even closer to Rozanov's limit remains a desirable goal. In Sec. II of the Supplemental Material [26], we show that both increasing the loss tangent of the dielectric or the surface resistance of the *MXene* enhance the absorbance of the MMA of a constant ϵ_r , whereas the absorbance of the optimized MMA is not enhanced. Since MMAs of a constant ϵ_r benefit from increasing dielectric and conductor losses, that may suggest that starting the optimization from a higher loss and higher absorbance MMA design may lead to an even more broadband optimized MMA. Furthermore, according to Ref. [31], creating MMA supercells composed of two or more pyramids of different patch lengths may also widen the total absorption bandwidth. The patches of each pyramid may offer high absorption in different parts of the spectrum. By properly selecting the geometric parameters of each pyramid, a wider absorption bandwidth may be achieved.

VI. FABRICATION AND EXPERIMENTAL CHARACTERIZATION

A. Fabrication of spatially variable dielectric layers

The dielectric layers are fabricated by using the Formlabs' Form3 SLA 3D printer. Clear V4 resin is used for all dielectric layers. Photographs of layers 1 to 6 of Fig. 15's Manufacturable MMA 8, captured by an AmScope MU1000 microscope, are shown in Fig. 16, where the detailed dielectric patterns are observed. We note that we noticed that small air holes are printed out somewhat smaller than resin parts of the same dimensions. We compensate for this fabrication aspect by increasing all air cell dimensions and correspondingly decreasing all resin cell dimensions prior to fabrication, so that the air cells are properly formed and are comparable to the resin cells.

B. Fabrication and characterization of the *MXenes*

We utilize *MXene* patches of thickness $t_{\text{MX}} = 0.9$ μm for the metal layers. The *MXene* and polycarbonate

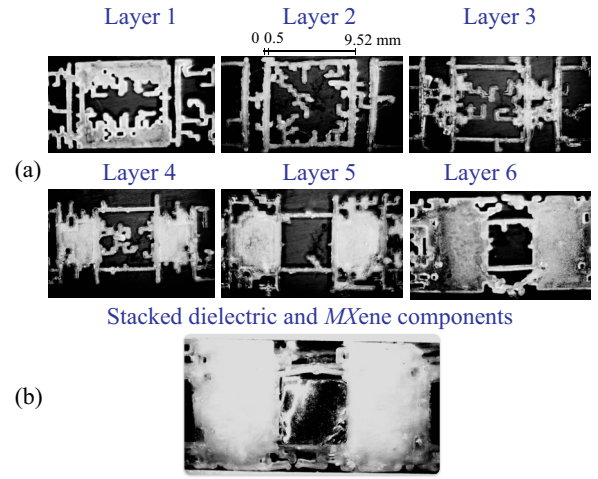


FIG. 16. Photographs of (a) the manufacturable spatially variable dielectric layers and (b) the top view of a stack of the dielectric layers and *MXene* patches, captured by an AmScope MU1000 microscope.

membrane samples were fabricated by vacuum-assisted filtration of $\text{Ti}_3\text{C}_2\text{T}_x$ aqueous dispersions (2 mg/ml in DI water). Specifically, 0.25 ml of dispersion per cm^2 of polycarbonate membrane was filtered and then the membranes were dried at room temperature for 2 h prior to use. We first estimate the bulk conductivity σ_{MX} in the X-band. We consider circular *MXene* samples with a radius of 1.7 cm deposited on a polycarbonate membrane of thickness $t_{\text{PC}} = 20$ μm and measure the reflection and transmission coefficients using a vector network analyzer (VNA) and WR90 waveguide components [32]. A through—reflect—line (TRL) calibration is performed before the measurement. As the *MXene*'s skin depth is much greater than its thickness, the surface resistance R_s is related to the bulk *MXene* conductivity and thickness by a quasistatic approximation $R_s = 1/(\sigma_{\text{MX}} t_{\text{MX}})$ [33]. We used a simple transfer-matrix method to estimate the value of the surface resistance R_s . We vary the surface resistance, and select the R_s value for which the calculated transmission coefficient is a closer match to the measured value.

We then cover the *MXene* samples with scotch tape to prevent them from becoming damaged and cut them into patches of appropriate dimensions. We stack the dielectric and *MXene* patches in a WR90 waveguide section of 4.95 mm thickness. The top view of the six stacked metal and dielectric layers is shown in Fig. 16, where we can also have a clear view of the top *MXene* patches. The remaining smaller subcomponents seven to nine are added on the top and sealed with scotch tape, to keep the stack intact. A double copper tape layer is also added just below the first (lowest) subcomponent to achieve metal backing.

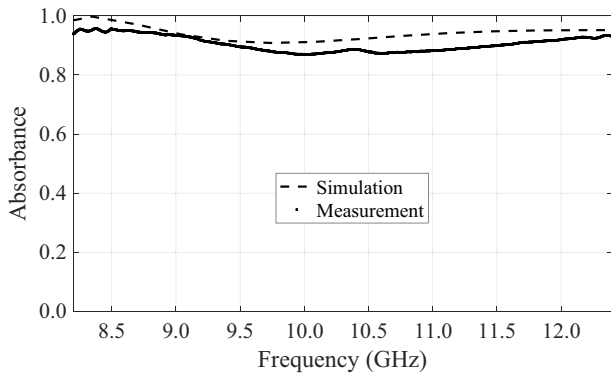


FIG. 17. Measured and simulated absorbance of the manufacturable multilayer MMA with spatially variable dielectric layers.

C. MMA measurement

The S parameters of the synthesized manufacturable MMA are measured in the X-band using VNA and WR90 waveguide-to-SMA adapters. A TRL calibration is performed before the measurement. The measured absorbance is compared to the corresponding FEM simulations in Fig. 17 and a satisfactory agreement is observed. Both the measurements and simulations of Fig. 17 correspond to the manufacturable MMA 8 in Fig. 15. Although the MMA operates in a much broader frequency range, we carried out measurements only in the X-band, as the MMA's lateral dimensions exactly match the WR90 waveguide's inner dimensions. We note that a minor mismatch between measurements and simulations is observed and may be attributed to the following reasons: (i) the MX ene patches were cut into the appropriate dimensions manually, so minor deviations from the target dimensions may affect absorbance, and (ii) the MX ene patches are set as a zero-thickness boundary condition in the simulations, whereas there is a finite thickness in the measurements. Specifically, the thickness is $t_{MX} + t_{PC} = 0.0209$ mm, plus a scotch tape layer of thickness $t_{TP} = 0.04$ mm, total 0.0609 mm, which we added on top of the MX ene, to prevent it from becoming damaged. The measurements serve as a proof of concept for the implementation of MMAs with spatially variable substrates and MX enes.

VII. CONCLUSION

We introduced a systematic approach for enhancing the absorbance of multilayer MMAs by combining spatially variable deep-subwavelength dielectric layers and lossy MX enes. MX enes offer increased Ohmic losses compared with PCBs and are thinner than conductive filaments. We demonstrate that synthesizing optimized spatially variable dielectric substrates instead of uniform ones can provide additional design flexibility and, in turn, enhanced absorbance. We inversely design an MMA with spatially

variable dielectric layers and obtain an optimized device with the dielectric constant having values in a continuous range. We developed the BINACONN3D methodology to transform the nonmanufacturable MMA into a manufacturable one, while preserving the performance. Fabrication and measurements in the X-band complement our synthesis process. Synthesizing optimized complex shapes both on the dielectric profiles and the MX ene layers may provide even higher design flexibility and pave the way toward approaching closer to Rozanov's limit. Our approach can be extended to different applications, which may also benefit from substituting existing uniform dielectric layers with spatially variable ones to enhance performance.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [34].

APPENDIX: FINITE-ELEMENT-METHOD SIMULATIONS

Finite-element-method (FEM) simulations are conducted in COMSOL Multiphysics. As MMA measurements will be carried out using WR90 waveguide components, throughout the paper we assume a computational domain with lateral dimensions corresponding to the inner dimensions of the WR90. The MMA is excited by a TE_{10} mode and perfect electric conductor (PEC) boundary conditions applied to the side boundaries and the ground. MX enes are introduced in the FEM simulations as boundary conditions of finite conductivity (transition boundary condition). The simulated absorbance A is calculated in terms of the S parameters S_{i1} , as $A = 1 - \sum_{i=1}^N |S_{i1}|^2$, where S_{11} is the reflection coefficient, S_{i1} , with $i > 1$, are the S parameters of the higher-order waveguide modes. Specifically, apart from the dominant TE_{10} mode, higher-order modes may be also excited as the frequency increases, such as TE_{20} , TE_{01} , TE_{11} , TM_{11} , TE_{30} , TE_{21} , TM_{21} , TE_{31} , and TM_{31} .

We consider two simulation environments: (i) one used for the MMAs with continuously varying dielectric properties. In this environment, we simulate the MMA of a constant ϵ_r and the optimized MMA with continuously varying dielectric properties. In this environment, there is no need to partition each dielectric substrate into a square grid. (ii) The second environment is used for the two, four,

and seven material MMA as well as the binary MMA simulations. In that case, we consider a cube grid with a cell size of approximately 0.5 mm and assign the necessary dielectric constant to each grid cell. In the first environment, we use a coarser mesh for the optimization, allowing it to be completed within a reasonably short timeframe. We then use a finer mesh to simulate the absorbance of the MMA of a constant ϵ_r and the optimized MMA. The coarser mesh in the first environment utilizes 79 162 first-order tetrahedral elements, resulting in 69 658 of freedom. For the inverse design, we use approximately 10 000 optimization variables. We optimize the absorbance over 25 frequencies within $f = [8, 25]$ GHz. The optimization process is completed in approximately 14 h on an AMD Ryzen Threadripper 3960X system (24 cores, 48 threads, 3.80 GHz, DDR4-3200). The finer mesh in the first environment utilizes 649 190 first-order tetrahedral elements, resulting in 571 244 of freedom. The FEM needs 46 s per frequency on the same computer. In the second environment, we use 311 564 first-order elements—tetrahedral for air and cubic for the MMA—resulting in 632 450 of freedom. The FEM needs 44 s per frequency on the same computer.

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