Complete Complex Amplitude Modulation with Electronically Tunable Graphene Plasmonic Metamolecules

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* Supporting Information

ABSTRACT: Dynamic high-resolution wavefront modulation of light is a long-standing quest in photonics. Metasurfaces have shown potential for realizing light manipulation with subwavelength resolution through nanoscale optical elements, or metaatoms, to overcome the limitations of conventional spatial light modulators. State-of-the-art active metasurfaces operate via phase modulation of the metaatoms, and their inability to also independently control the scattered amplitude leads to an inferior reconstruction of the desired wavefronts. This fundamental problem posed severe performance limitations particularly for applications relying on subwavelength spatiotemporal complex field modulation, which includes dynamic holography, high-resolution imaging, optical tweezing, and optical information processing. Here, we present the “metamolecule” strategy, which incorporates two independent subwavelength scatterers composed of noble metal antennas coupled to gate-tunable graphene plasmonic nanoresonators. The two-parametric control of the metamolecule secures the complete control of both amplitude and phase of light, enabling $2\pi$ phase shift as well as large amplitude modulation including perfect absorption. We further develop a generalized graphical model to examine the underlying requirements for complete complex amplitude modulation, offering intuitive design guidelines to maximize the tunability in metasurfaces. To illustrate the reconfigurable capability of our designs, we demonstrate dynamic beam steering and holographic wavefront reconstruction in periodically arranged metamolecules.

KEYWORDS: graphene, plasmonics, nanoresonator, mid-infrared, complex amplitude modulation

Wavefront engineering is a crucial technology for holography, high-resolution imaging, optical tweezing, optical data storage, and communication. In the past decade, metasurfaces have emerged as an effective means of engineering optical wavefronts. They have been used to produce nonintuitive refraction phenomena and high-resolution images beyond the diffraction limit, all of which are otherwise unachievable in conventional optical devices. Metasurfaces work by utilizing subwavelength scatterers, i.e., metaatoms, to locally control the amplitude and phase of reflected or refracted light. In most cases, those metaatoms are composed of a metal or dielectric structure with one or more scattering resonances near the operating frequency. The use of metasurfaces to modulate the amplitude and phase of the scattered light has enabled general wavefront manipulation from ultrathin and flat surfaces. Dynamic reconfigurability of a metasurface is highly desirable, as it would extend the application space to holographic projections, fast optical beam steering or routing, and high-speed aberration correction in microscopes or telescopes. To impart dynamic tunability in these devices, materials with tunable optical properties are included in the scattering structures, which allows the scattering resonances to be shifted in frequency and amplitude, which in turn affects their scattering behavior. For example, by including graphene or indium tin oxide (ITO), tunable metasurfaces have been experimentally demonstrated that can act as either a perfect

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Similar structures have been used to realize dynamic beam steering\textsuperscript{16} and to continuously control the phase of a reflected light beam up to $1.2\pi$ and $1.7\pi$ for graphene\textsuperscript{17} and ITO,\textsuperscript{18} respectively. Likewise, direct-gap semiconductors,\textsuperscript{19} thermo-optically tuned silicon,\textsuperscript{20} and other phase change materials\textsuperscript{21,22} have also been utilized to dynamically change the scattering behavior of subwavelength structures.

Despite the rapid progress in the field, current dynamic metasurfaces suffer from an inevitable interference between amplitude modulation and phase modulation, where the change of scattered phase also unavoidably alters its scattered amplitude, and vice versa. Furthermore, no device thus far has been demonstrated to dynamically tune a scattered wavefront over a $2\pi$ of phase space. These problems stem from the basic tuning mechanism. Current metasurfaces have been designed based on the regulation of a single resonance in an individual metaatom, which limits the degrees of freedom to control two components independently, i.e., the amplitude and the phase of light, and the access to the complex amplitude space. The
limited tunability results in lowering the resolution of images or creating undesirable noise in images. To the best of our knowledge, no active metasurface has yet been reported that can control both phase and amplitude independently, i.e., complex amplitude modulation, which is necessary for complete wavefront reconstruction.

In this paper, we present an electronically reconfigurable, reflecting metasurface structure composed of subwavelength scattering elements that can locally alter the phase of scattered light over a complete 2π phase coverage, in conjunction with independent control of the amplitude of the scattered light. The unit cell of our proposed structure, herein denoted as metamolecule, consists of a pair of noble metal antennas, each in contact with graphene plasmonic ribbons (GPRs). Here, the pair of Fermi levels of the GPR constitute the two control parameters for our metamolecule in modulating the amplitude and the phase of light. Each metamolecule operates at 7 μm and is subwavelength, such that only their collective responses are observable in the far field. In other words, this metamolecule configuration composes assembled optical responses from a pair of metaatoms, which displays entirely different optical behaviors from each metaatom and provides more controllability in complex amplitude modulation. In addition, a generalized graphical approach based on effective surface admittance elucidates the underlying preconditions for the complete complex amplitude modulation, providing intuitive design guidelines to maximize the tunability of the metamolecules. To illustrate the reconfigurable capability of our designs, we demonstrate dynamic beam steering and holographic wavefront reconstruction in a structurally identical metasurface by simply tuning the graphene Fermi levels of the metamolecules.

RESULTS AND DISCUSSION

Device Geometry and Complex Amplitude Modulation. Graphene is an exotic mid-infrared plasmonic material, with extremely confined plasmonic modes that are highly electronically tunable due to its atomic thickness.9–13 In addition, the small optical-mode volumes of graphene plasmons are beneficial for miniaturizing the active plasmonic devices as well as driving strong light–matter interactions in a deep subwavelength scale. These optoelectronic properties of graphene allow for the creation of electronically tunable light modulation devices controlling the intensity and phase of light from the mid-infrared to THz. In particular, our recent experimental results show that the key components of our proposed design are experimentally feasible; namely, the strong light–matter interactions that occur between noble metal antennas and GPRs have been shown to enable large dielectric tunability.13,14 To be specific, it was demonstrated that oscillator strength in GPRs can be also significantly enhanced by incorporating multiscale nanophotonic structures consisting of noble metal plasmonic structures.13,14,31–40 These subwavelength noble metal plasmonic structures improve the radiative coupling to the GPRs by bridging the large wavelength mismatch between free space photons and deep-subwavelength scale graphene plasmons. In addition, nonresonant field enhancement in the noble metal plasmonic structures augments the oscillator strengths in the GPRs, enabling a large tunability in the local dielectric environment. It will be shown that the incorporation of multiscale nanophotonic structures allows our metamolecule structure to display large amplitude modulation and 2π phase shift, even with an assumed graphene carrier mobility of ≤1000 cm² V⁻¹ s⁻¹, which is readily accessible in CVD-grown graphene.41

Figure 1a shows the proposed metasurface consisting of periodically arranged graphene plasmonic metamolecules, modulating the complex amplitude of the reflected mid-infrared light. The GPRs are combined with noble metal plasmonic structures to enhance the tunability, and a back reflector is incorporated to gate the graphene electrostatically and to further intensify the light–matter interactions in the graphene.13,38,39 In a single metamolecule, or a unit cell of the metasurface, a pair of graphene plasmonic metaatoms are arranged side by side. The GPR is asymmetrically placed on one side of a metal strip such that each GPR is electronically isolated from another metal strip. This configuration allows us to tune the graphene Fermi level in each metaatom individually through a gating voltage between the back gate and the metal strip connected to the GPR, thus providing two degrees of freedom necessary for independently modulating the amplitude and the phase of the reflected light. In this way, a metasurface consisting of periodically arranged graphene plasmonic metamolecules enables the electronic modulation of the complex amplitude by simply tuning the two graphene Fermi levels of the metamolecule. Here, we point out that the dynamic optical resonances in the proposed metasurface originate from strong plasmonic resonances in the GPRs, and not the metal strips. In fact, the plasmonic resonances of the metal strip are very much detuned from that of graphene. The metal strips are exploited to enhance the oscillator strengths of the GPRs by nonresonant focusing effects42 and do not support additional plasmonic resonances. This implies that an optical response of each metaatom is independently tunable with negligible intercoupling between the adjacent metaatoms.

Figure 1b,c illustrate the amplitude and the phase of reflection coefficients of the proposed structures as a function of the graphene Fermi levels (E_F1 and E_F2, as denoted in Figure 1a). The geometries of the structure are carefully tailored to exhibit maximum tunability at a free space wavelength of 7 μm with a graphene carrier mobility of 1000 cm² V⁻¹ s⁻¹, and normal incoming light with transverse magnetic polarization is assumed. The structural parameters are presented in the caption of Figure 1a. Figure 1d,e illustrate the capabilities of the complex amplitude modulation in the proposed metamolecules. A 2π phase shift with a fixed amplitude (|r_l| = 0.5) of reflection and a large amplitude modulation with a fixed phase (ϕ = 0) metaatom can be achieved through the two-parametric tuning, E_F1 and E_F2, of the metamolecules. Representative electric field distributions around the metamolecules are displayed in Figure 1f,g.

Figure 1b shows that two perfect absorption conditions exist at different ranges of graphene Fermi levels, whereas perfect absorption is not achievable in a single metaatom. The graphene Fermi levels in Figure 1d,e are chosen along the solid blue line (constant amplitude, |r_l| = 0.5) and the solid red line (constant phase, ϕ = 0) in Figure 1b,c, respectively. The graphene Fermi levels (E_F1 and E_F2) used in Figure 1d,e are summarized in the Supporting Information, Table S1. Similar complex amplitude modulations can also be realized along the dotted lines in Figure 1b,c and the calculation results are presented in the Supporting Information, Figure S1.

Generalized Graphical Approach to Complex Amplitude Modulation. Admittance analysis is convenient for investigating a stratified structure by a series of admittance for
each layer. Moreover, our previous results demonstrated that effective surface admittance accurately describes graphene plasmonic nanostructures when the thickness and the period of the structures are in the subwavelength regime. To elucidate the underlying requirements for the complete complex amplitude modulation, we develop a generalized graphical model based on surface admittance. This approach can be exploited to devise general metasurfaces and to analyze their dynamic complex amplitude behaviors.

The proposed structure can be divided into two parts: a structured surface and a substrate including a back reflector. Then, the complex reflection coefficient of the structure in terms of the admittances with normal incident light is derived as

\[
|r| = \frac{\tilde{Y}_s + \tilde{Y}_{\text{sub}} - 1}{\tilde{Y}_s + \tilde{Y}_{\text{sub}} + 1}
\]

Figure 2. (a) Surface admittances and (b) complex amplitudes in the ideal case exhibiting closed circles. (c) Surface admittances and (d) complex amplitudes in an actual case exhibiting arcs. In (a)–(d), the solid color lines correspond to the metaatoms, and the shaded areas represent the reconstructed area by the metamolecule. The dotted lines correspond to the denoted amplitudes (|r|), and the gray lines in (a) and (c) represent the negative imaginary part of the substrate admittance (\(\tilde{Y}_{\text{sub}}\)). In (d), the maximum amplitude covering 100% of the area is 0.566.
Here, the substrate admittance can be considered as a purely imaginary value when there is negligible absorption in the substrate. More details on the surface admittance model are given in the Supporting Information, Note S1.

The advantage of this approach is that we can easily ascertain the surface admittances necessary for a specific complex amplitude modulation range by using eq 1. For example, by transforming the origin of the \( r \)-plane, we can obtain the required surface admittance condition, \( \tilde{Y}_s = 1 - \tilde{Y}_{\text{sub}} \), for perfect absorption \((r = 0)\). We point out that any set of concentric circles centered at the origin of the \( r \)-plane \((i.e., \text{the conditions for phase modulation with constant reflection amplitudes})\) can be mapped onto a set of nonconcentric circles with radii of \( 2|r|/(1 - |r|^2) \), as displayed by the dotted lines in Figure 2a (see Supporting Information, Note S2). In particular, all centers of the dotted lines are placed on the \(-\text{Im}(\tilde{Y}_{\text{sub}})\) axis.

Recognizing that the surface admittance of a graphene plasmonic metasurface inherently spans a circular trajectory in the \( \tilde{Y}_s \)-plane, the problem of designing its dynamic response can be reduced to locating the desired center and radius of the circle in the \( \tilde{Y}_s \)-plane by judiciously choosing the structural parameters of the metasurface.

The dynamic behavior of the metamolecule can be deduced from the properties of the constituent metaatoms. The solid red \((\tilde{Y}_{s1})\) and blue \((\tilde{Y}_{s2})\) curves in Figure 2a correspond to the proposed metaatoms’ surface admittances in the ideal case. Note that the centers of both circles are aligned with the \(-\text{Im}(\tilde{Y}_{\text{sub}})\) axis to maximize the tunability of the complex amplitude. Since the two types of metaatoms are connected serially, the effective surface admittance \( \tilde{Y}_s \) of the metamolecule is obtained by the weighted harmonic mean of the two surface admittances as \( \tilde{Y}_s^{-1} = (p_1 \tilde{Y}_{s1}^{-1} + p_2 \tilde{Y}_{s2}^{-1})/(p_1 + p_2) \). In the ideal case where \( \tilde{Y}_{s1}(E_{F1}) \) and \( \tilde{Y}_{s2}(E_{F2}) \) form closed circles, the resulting surface admittance of the metamolecule \( \tilde{Y}_s(E_{F1}, E_{F2}) \) spans the disc area displayed as the shaded region in Figure 2a. In other words, by tuning the graphene Fermi levels of the two metaatoms, one can independently control the amplitude and the phase of the reflection from the metamolecule within the shaded area in Figure 2b. In Figure 2a,b, small voids appear inside the reconstructed areas of the metamolecule. These voids originate from the weighted averaging method, and we can eliminate the voids by balancing the radii of the \( \tilde{Y}_s \)-circles and the pitches of the metaatoms (see Supporting Information, Note S3).

In realistic devices, the range of graphene Fermi level is bounded by the breakdown voltage of the insulating layer. Due to the limited tuning range, the surface admittance curves of the metaatoms cannot produce a closed circle but lead to the formation of an arc with a small opening as displayed by solid color lines in Figure 2c. Here, we assume that the graphene

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**Figure 3.** (a) Schematic of the circuit model for a metaatom. Surface admittances evaluated by the circuit model (b) as the pitch is varied with a fixed metal gap and (c) as the metal gap is varied with a fixed pitch. In (b) and (c), the solid color lines are calculated by full-wave simulations, and the dotted color lines are evaluated by the circuit model.
Fermi level can be tuned from 0 to 1 eV. This limitation causes additional unfilled areas in the reconstructed surface impedance area (see Supporting Information, Note S3). The surface admittance lines stop extending at the end due to the limited range of the graphene Fermi levels. In addition, the interband absorption in graphene inherently produces small deviations at low graphene Fermi levels, resulting in small bending. In spite of these imperfections, Figure 2c shows that the metamolecule covers a wide range of surface admittance values. The dotted color lines indicate the surface admittances corresponding to the various reflection amplitudes.

Figure 2d shows the reconstructed complex amplitudes from the metamolecule (the shaded area) by independently tuning the metaatoms (the solid color lines). The proposed metamolecules produce 100% of the $|r| \leq 0.566$ domain and 98.1% of the $|r| \leq 0.6$ domain. In particular, Figure 2d illustrates the limited modulation range when we rely on regulating a single resonance in metasurfaces. The results given in Figure 2d are calculated by full-wave simulations. In the

Figure 4. Beam steering with angles ($\theta_r$) of (a) 20.9°, (b) 45.5°, and (c) 72.0°. In (a)–(c), the target amplitudes are fixed at 0.5, and the scale bars are 5 μm. (d) Magnitude of the scattering parameters ($|S|$) of the beam steering. (e) Schematic of holographic wavefront calculation and reconstruction with metamolecules. (f) Reflected magnetic intensity ($|H_f^2/H_0^2|$) distribution of single focusing with a beam spot located at $(x, z) = (0 \mu m, 7 \mu m)$, where a free space wavelength ($\lambda_0$) is 7 μm. Here, the air/metasurface boundary corresponds to $z = 0$. The maximum intensity at the focal spot is 2.21. (g) Magnetic intensity ($|H_f^2/H_0^2|$) distribution of triple focusing with beam spots located at $(x_1, z_1) = (-14 \mu m, 56 \mu m)$, $(x_2, z_2) = (0 \mu m, 31 \mu m)$, and $(x_3, z_3) = (14 \mu m, 56 \mu m)$. The maximum intensities and the transverse full width half-maximums (FWHMs) at the focal spots are 1.48 and 0.77$\lambda_0$ for $\bullet$, 1.57 and 0.43$\lambda_0$ for $\bigcirc$, and 1.46 and 0.50$\lambda_0$ for $\odot$. In (f) and (g), the metasurface consists of 75 metamolecules, and the scale bars are 20 μm.
Supporting Information, Figure S7 reveals that the evaluation by the weighted averaging method for the metamolecule admittance shows excellent agreement with the full-wave simulation results. This comparison also confirms that the intercoupling between the metaatoms is negligible. More details are presented in the Supporting Information, Note S4.

**Circuit Analysis.** To maximize the tunability in the complex amplitude plane, the metamolecule should be designed such that the surface admittance curves of the constituent metaatoms have maximum radii, while their centers are placed on the $-\text{Im}(\hat{Y}_{\text{sub}})$ axis. To understand the electromagnetic behaviors depending on the structural parameters, we develop an analytic model describing the metasurface as a collection of effective circuit components, as shown in Figure 3a. The normalized surface admittance of a metasurface composed of a single metaatom can be expressed as

$$\hat{Y}_s = \hat{Y}_{\text{metal}} + \frac{p}{g} \hat{Y}_{\text{GPR}}$$

$$= \frac{p}{Y_0} \left[ -i \omega (L_m + L_k) + \frac{1}{-i \omega (C_c + C_g)} \right]^{-1} + \frac{p}{g} \hat{Y}_{\text{GPR}}$$

(2)

where $\omega$, $Y_{\text{metal}}$, and $Y_{\text{GPR}}$ are the operating frequency, normalized surface admittance of the metallic structure, and normalized surface admittance of the equivalent GPR array without the metallic structure, respectively. The elemental inductance ($L$) and capacitance ($C$) are determined by the structural parameters, and $L_{\text{metal}}$, $L_k$, $C_c$, and $C_g$ correspond to the magnetic inductance, kinetic inductance, coplanar capacitance, and gap capacitance of the metallic structure, respectively. More details are provided in the Supporting Information, Note S5. We note that the analytic circuit model shows good agreement with full-wave simulation results as shown in Figure 3b,c. The advantage of this circuit model is that we can investigate the electromagnetic behaviors of the GPRs and the metallic structure separately. The $(p/g)\hat{Y}_{\text{GPR}}$ in eq 2 describes the plasmonic resonances of the GPRs depending on the graphene Fermi level and thus determines the radius of the surface admittance curve. Here, the $p/g$ ratio is the field enhancement factor originating from the nonresonant antenna effect by the metallic structure. A larger $p/g$ ratio enhances the oscillator strength of the GPRs, resulting in a larger surface admittance circle, as shown in Figure 3a. The radius of the oscillator strength of the GPRs, resulting in a larger surface admittance circle depends linearly on $1/g$, but quadratically on $p$ because both the field enhancement factor and the density of the equivalent GPR array are directly proportional to $1/g$. The widths ($w$) of the GPRs primarily determine the resonant graphene Fermi level of a metaatom. In our proposed device, the GPR widths of the two metaatoms are chosen to be 45 and 50 nm. These GPR widths lead to plasmonic resonances at the graphene Fermi levels of 0.506 and 0.603 eV, respectively, at a free space wavelength of 7 $\mu$m (see Supporting Information, Figure S3).

The imaginary offset of the surface admittance curve is mainly determined by the $\hat{Y}_{\text{metal}}$ in eq 2. In our design space, the $\hat{Y}_{\text{metal}}$ is dominated by the capacitance of the metallic structure, whose contribution to the surface impedance is an order of magnitude higher than that of the inductive components. For a given thickness of the metallic layer ($h$), which in our case is approximately 2 orders of magnitude shorter than the free space wavelength to ensure effective surface description, the imaginary offset increases in the negative direction (i.e., the metasurface becomes more capacitive) with larger pitches ($p$) and with narrower metallic gaps ($g$), as shown in Figure 3b,c. Since $C_c$ and $C_g$ are proportional to $\log(p/g)$ and $1/g$, respectively, $\hat{Y}_{\text{metal}} \approx -i \omega (C_c + C_g)/Y_0$ depends superlinearly on $p$ and sublinearly on $1/g$. For both metaatoms, the pitches and the gaps are chosen to align the $\hat{Y}_r$-circles on the $-\text{Im}(\hat{Y}_{\text{sub}})$ axis.

**Active Beam Steering and Holographic Wavefront Reconstruction.** Figure 4 illustrates the capability of the proposed metamolecules in dynamic wavefront reconstruction: active beam steering and beam focusing via holographic wavefront reconstruction. In these demonstrations, the metasurfaces are composed of periodically arranged meta-atoms, and the graphene Fermi levels are tuned to generate target complex amplitudes.

In active beam steering, the graphene Fermi levels are tuned to generate a linearly graded phase profile, $\phi(x) = k_x x$ with a constant amplitude of $|\psi| = 0.5$, where $k_x$ is the in-plane wavevector of the target output beam. Since the width of a metamolecule, $p = p_1 + p_2 = 2454$ nm, is much shorter than the free space wavelength $\lambda_0 = 7$ $\mu$m, the proposed device can sufficiently resolve the wavefront of a beam with a large deflection angle. Therefore, unlike conventional spatial light modulators whose elements are larger than the diffraction limit, this device can steer a light beam at a wide angle without the aid of external magnifying optics. As shown in Figure 4a–c, the full-wave simulation results demonstrate that the reflected wavefronts are well aligned with the target refraction angles of 20.9°, 45.5°, and 72.0°, which correspond to the phase ramp $\Delta \phi = k_x p = \pi/4, \pi/2, \text{and } 2\pi/3$, respectively. The scattering parameters $S_11$ presented in Figure 4d confirm good beam steering performance with small side lobes, where nonzero side lobes would originate from discrete sampling of the phase front and crosstalk between the neighboring metamolecules.

Figure 4e displays a schematic of the holographic wavefront calculation and reconstruction for beam focusing. By placing sources at desired positions and evaluating the complex amplitude of light propagating from the sources, we can analytically calculate the amplitude and the phase of light along a metasurface plane. After tuning the graphene Fermi levels to produce the necessary complex amplitudes, we can reconstruct the focal points in the free space. Figure 4fg show light focusing with a single spot and triple spots with 75 metamolecules via full-wave simulations. The target amplitude $|\psi|$ and the phase $\phi$ profiles used for these calculations are presented in the bottom panels. The subwavelength wavefront resolution of the metamolecules, combined with their ability to address arbitrary complex amplitude response, makes possible the generation of a clear and compact focal spot merely a single wavelength above the metasurface, as shown in Figure 4f. The transverse and vertical full width at half-maximums (FWHMs) of the focal spot are approximately 0.36$\lambda_0$ and 0.77$\lambda_0$, respectively, indicating high-resolution wavefront reconstructions. As the focal length increases, the size of the focal spot becomes slightly larger due to the decreased numerical aperture, but the field intensity at the focal point increases, as illustrated in the Supporting Information, Figure S10. The same metasurface can also produce multiple focal spots in free space via the same methodology prescribed earlier. As shown in Figure 4g, the metasurface can construct rapidly varying phase and amplitude profiles for three focal spots separated by
a few wavelengths from each other, while maintaining the transverse FWHM of the foci to be under $0.52 \lambda_0$. These high-resolution focal spots can only be produced when both the amplitude and phase of light are precisely controlled, a clear advancement that our metamolecule-based metasurface has achieved.

Dependency on Graphene Carrier Mobility. The structural parameters of the metamolecule presented in Figure 1a are designed to exhibit maximum tunability with a graphene carrier mobility ($\mu$) of 1000 cm$^2$ V$^{-1}$ s$^{-1}$, and the maximum graphene Fermi level ($E_{F,\text{max}}$) is assumed to be 1 eV. In experiments, these conditions might not be accessible. First, inevitable imperfections in the processed graphene samples, such as edge defects in the GPRs and residues, lower the effective graphene carrier mobility. In our previous research, we obtained $\mu \approx 500$ cm$^2$ V$^{-1}$ s$^{-1}$ in patterned graphene. In addition, $E_{F,\text{max}}$ is limited by the breakdown voltage of the insulating layer, and $E_{F,\text{max}} \approx 0.6$ eV for a normal dielectric layer, such as SiO$_2$ or SiN$_x$, using an electrostatic gating method.

In spite of these limitations, complete complex amplitude modulation is still achievable in the proposed metamolecules by tuning the structural parameters. Complex amplitude modulations with different graphene carrier mobilities are presented in Figure 5. The geometries used in Figure 5 are adjusted to exhibit the maximum modulation range for the corresponding graphene carrier mobility when we assume that $E_{F,\text{max}}$ is 0.6 eV. The structural parameters are summarized in
the Supporting Information, Table S3. Of course, a higher $\mu$ and a larger $E_{F,\text{max}}$ enlarge the range of complex amplitude modulations owing to stronger oscillator strengths in the GPRs, as shown in Figure S. However, this relationship does not indicate that a complete complex modulation is infeasible with low graphene carrier mobilities and a limited range of graphene Fermi levels. As demonstrated in Figure 5a, a complete complex amplitude modulation is achievable within the range of $|r| = 0.186$ even with $\mu = 500$ cm$^2$ V$^{-1}$ s$^{-1}$ and $E_{F,\text{max}} = 0.6$ eV.

**CONCLUSIONS**

In summary, graphene plasmonic metamolecules enable complete complex amplitude modulation at mid-infrared frequencies. In the proposed metamolecules, electronically tunable responses are achieved by graphene plasmonic ribbons, and noble metal plasmonic structures are incorporated to enhance the light–matter interactions. In particular, two degrees of freedom required to independently modulate the amplitude and phase of light are obtainable through individual control of the graphene Fermi levels of the two constituent metasurfaces. To investigate the electromagnetic behaviors of the metamolecules, we developed a graphical method based on the surface admittance and surface impedance. This approach offers intuitive guidelines for designing metamolecules as well as general metasurfaces for complex amplitude modulation. In addition, the proposed circuit model allows the efficient design of metamolecules without involving brute-force simulations. The capabilities of the proposed metamolecules were proven by demonstrating active beam steering and holographic wavefront reconstruction with high figure-of-merit. Our proposed metamolecule represents a conceptual advancement to metasurface design, allowing for complete amplitude and phase control of light, and should find its application in thermal engineering, optical phased arrays, and real-time hologram systems beyond the mid-infrared.

**METHODS**

**Optical Simulations.** The frequency-dependent dielectric functions of SiO$_2$ and Au were obtained from the Palik data.$^{43}$ Graphene was modeled by a sheet with zero thickness, and its optical conductivity $\sigma$ was calculated by local random phase approximation,$^{14,45}$ which depends on the frequency $\omega$ and the graphene Fermi level $E_g$ as follows:

$$\sigma(\omega, E_g) = \frac{ie^2 \nu_F T}{\pi \hbar^2 (\omega + i\Gamma)} \log \left[ 2 \cosh \left( \frac{E_g}{2k_B T} \right) \right] + \frac{ie^2 (\omega + i\Gamma)}{\pi} \times$$

$$\int_0^\infty \frac{dE}{(\hbar^2 (\omega + i\Gamma) - (2E)^2)}$$

where $f(E) = 1/(1 + \exp[(E - E_g)/k_B T])$, $\Gamma = \varepsilon_0 \nu_F /\mu E_g$ is the scattering rate, $\nu_F$ is the Fermi velocity, and $\mu$ is the graphene carrier mobility. The temperature $T$ was set as 300 K. The optical responses of the metasurfaces and the metamolecules were calculated by full-wave simulations with the finite-difference time-domain method and the finite element method, and the results from both simulation methods were crosschecked for validation.

**Effective Surface Admittance.** A thin metasurface that has a deep subwavelength thickness can be modeled by a two-dimensional sheet with an effective surface admittance ($Y_s$). The surface admittances of the metasurfaces were evaluated from the given reflection coefficients ($r$) by $Y_s = (1 - r)/(1 + r) - Y_{sub}$ where $Y_{sub}$ is the substrate admittance. The effective surface impedance ($Z_s$) of a metasurface can be directly calculated from its surface admittance by $Z_s = Y_s^{-1}$. Using this model, the optical properties of the full metasurface stacks can be predicted analytically by solving a set of simple equations. In the quasi-static limit, the surface admittance of a metasurface can be approximated by treating the metasurface as a connected set of lumped circuit elements, as described in the Supporting Information, Note S3. More details on the surface admittance and the surface impedance are presented in the Supporting Information, Notes S1–S3.

**ASSOCIATED CONTENT**

3 Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsnano.9b09277.

Details on surface admittance; relation among reflection coefficient, surface admittance, and surface impedance; analysis of reconstructed surface impedance area; details on circuit analysis; comparison between full-wave simulation results and analytic evaluation (PDF)

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*S. Han and Se. Kim contributed equally to this work. S.H. and M.S.J. conceived the ideas. S.H. and Se.K. performed the simulations and theoretical analysis. All authors co-wrote the paper, and Se.K. and M.S.J. supervised the project.

**Notes**

The authors declare no competing financial interest.

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