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Masyukov, Maxim S.; Grebenchukov, Alexander N.

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Temporally modulated metamaterial based on a multilayer graphene structure

Maxim S. Masyukov

Department of Electronics and Nanoengineering, Aalto University, P.O. Box 15500, 00076 Aalto, Finland

Alexander N. Grebenchukov

Tydex LLC, Domostroitel'naya 16, St. Petersburg 194292, Russia

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The recently developed concept of time-varying metamaterials paves the way for advanced methods of electromagnetic property modulation, such as magnet-free isolators, frequency converters, and other nonreciprocal photonic components. Here, we analyze the metamaterial based on the periodically arranged temporally modulated graphene layers via a steplike graphene Fermi level. The material properties are described by the effective medium theory. The electromagnetic wave temporal scattering phenomenon caused by rapid steplike graphene Fermi level switching is considered. It is demonstrated that the frequency of scattered waves increases as the Fermi level grows rapidly and vice versa. The relation between the amplitudes of forward- and backward-scattered waves is found using the Laplace transform technique. The proposed concept allows us to efficiently generate frequency harmonics under instant switching of the graphene Fermi level at a few meV. Our investigation provides a basement for the realization of temporally modulated multilayer structures based on functional tunable materials as power-efficient frequency converters.

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I. INTRODUCTION

Metamaterials and their planar equivalent metasurfaces are artificially engineered materials consisting of spatially distributed subwavelength scattering particles that exhibit exceptional optical properties [1]. However, most time-invariant metamaterials are static in nature. Their properties are fixed during the fabrication, which limits their application to only one specific function. The standard way to achieve postfabrication tunability is by introducing the reconfigurable part into metamaterial. The electromagnetic properties of that reconfigurable part can be tuned in a real-time manner by exploiting external stimuli [2]. On the other hand, this class of tunable metamaterials has been studied in a quasistatic regime without consideration of temporal variations.

Besides the commonly used spatial modulation in metamaterials, ultrafast material modulation in the time domain provides an additional degree of freedom, enabling us to tailor the electromagnetic response of media with emerging new wave phenomena [3–7]. Among such fascinating phenomena, magnetless nonreciprocity [8,9], nonreciprocal beam steering and focusing [10], Casimir light and two-photon emissions [11,12], intensity-independent frequency conversion [13,14] and harmonics generation [15,16], and broadband cloaking [17,18] and sensing [19] seem to be the most promising. Moreover, depending on the specific space-time modulation function, the multifunctional nonreciprocal metasurface can be realized [20,21].

In addition to these impressive phenomena, various types of dualities between temporal and spatial modulations have

been demonstrated, such as antireflection temporal coatings [22], the temporal Brewster angle [23], temporal beam steering [24], and the temporal counterpart of the Wood anomaly [25].

Electromagnetic phenomena in time-varying systems are well developed for the magnetoplasma medium [26]. Recently, inspired by such an approach, it was shown that a rapid decrease [27] or increase [28] in the graphene carrier density leads to plasmon frequency downshifting or upshifting, correspondingly, without plasmon amplification. Motivated by these outstanding opportunities, in this paper, we employ an effective medium approach to study the temporal scattering problem for a graphene-dielectric multilayered structure. In particular, we demonstrate that the frequency upshifting or downshifting of scattered waves is obtained by instantly changing the graphene Fermi level, depending on the sign of the Fermi level alteration independent of its initial value. Moreover, we establish the relation between amplitudes of scattered forward-propagating and back-propagating waves. In addition, we discuss practical implementation opportunities for the proposed temporally modulated multilayered structure.

This paper is organized as follows. In Sec. II, we describe the material parameters for the multilayered medium based on graphene and our main assumptions. In Sec. III, we analyze the frequency conversion in the temporally modulated medium and find the dependence of the scattered frequencies on the Fermi level change. Then, in Sec. IV we determine the amplitudes of scattered waves, perform the stability analysis, and study the spectral characteristics of Gaussian pulses in such a medium. After discussing possible realizations of this metamaterial in Sec. V, we conclude our work in Sec. VI and provide derivations of the equations in the Appendix.

*grebenchukov_a@mail.ru

FIG. 1. A schematic image of the multilayered graphene meta-material and its Fermi level under a steplike (a) Fermi level increase and (b) decrease.

II. MULTILAYERED GRAPHENE STRUCTURE AND BASIC EQUATIONS

The medium discussed in this paper is formed by graphene sheets with infinitely small thickness, aligned in the xy plane and separated by dielectric spacers with thickness d along the z axis. These conductive sheets are described by surface conductivity $\sigma(\omega)$, whereas the dielectric spacer is defined by the permittivity ε_d . Time dependence of the fields in the form $e^{j\omega t}$ is assumed. Here, we use a simplified Drude-like model for graphene surface conductivity [29]:

$$\sigma(\omega, \mu_F) = -\frac{je^2}{\pi\hbar^2} \frac{\mu_F}{\omega}, \quad (1)$$

where e is the elementary charge, $\hbar$ is the reduced Plank's constant, μ_F is the Fermi level, and ω is the angular frequency. Such a model avoids the impact of temperature effects, as well as electron collisions, and is valid for highly doped graphene. The electron collisions' influence on the fields after the rapid carrier density increase is small enough to consider [28]. A schematic representation of the medium is shown in Fig. 1.

The exact material parameters for the medium can be obtained by the implementation of the transfer matrix approach and Floquet-Bloch theory [30,31]. However, here, we assume the periodicity of the structure is extremely small in comparison to the operational wavelength [32]. Let us take one unit cell with the graphene sheet at $z = 0$ between two dielectric layers with a thickness of $0.5d$. Within this unit cell, the electric ($\mathbf{E}$) and magnetic ($\mathbf{H}$) fields are connected as [30]

$$\text{rot}\mathbf{H} = \mathbf{J} + \varepsilon_0\varepsilon_d \frac{\partial\mathbf{E}}{\partial t} = \mathbf{J} + j\omega\varepsilon_0\varepsilon_d\mathbf{E}, \quad (2a)$$

$$\mathbf{J} = \sigma\mathbf{E}_t\delta(z), \quad (2b)$$

where $\delta(z)$ is Dirac delta function, $\mathbf{E}_t$ is the tangential component of the electric field, and $\mathbf{J}$ is the surface current along the sheet. After integrating over the volume of one unit cell, we get the effective medium parameters for the dielectric

FIG. 2. Dependence of the tangential dielectric permittivity on the Fermi level values of graphene. The following parameters are used: $d = 1 \mu\text{m}$ and $\varepsilon_d = 4$, which is equivalent to using hexagonal boron nitride as a spacer [31].

permittivity tensor $\hat{\varepsilon}$ components:

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_t & 0 & 0 \\ 0 & \varepsilon_t & 0 \\ 0 & 0 & \varepsilon_d \end{pmatrix}, \quad (3)$$

where

$$\varepsilon_t = \varepsilon_d - j \frac{\sigma(\omega)}{d\omega\varepsilon_0} = \varepsilon_d - \frac{e^2\mu_F}{\pi d\hbar^2\omega^2\varepsilon_0}. \quad (4)$$

The typical dependency of the tangential effective permittivity on the Fermi level is shown in Fig. 2.

For simplicity, the effective plasma frequency of the medium is introduced as follows:

$$\omega_p^2 = \frac{e^2\mu_F}{\pi d\hbar^2\varepsilon_0} = \gamma \frac{\mu_F}{d}, \quad (5)$$

where γ is a proportionality coefficient between the plasma frequency, Fermi level, and periodicity. In this formalism, the effective conductivity of the medium can be rewritten in the following way:

$$\sigma_e = \frac{\sigma(\omega)}{d} = -j\varepsilon_0 \frac{\omega_p^2}{\omega}. \quad (6)$$

The approximations made here are identical to the Maxwell-Garnett effective medium approach for a hyperbolic metamaterial [33], and this model does not take into account the periodicity of the structure. If the periodicity is not small enough compared to the wavelength, more accurate models are needed [34].

III. FREQUENCY CONVERSION OF SCATTERED WAVES

Due to the multilayered structure of the metamaterial, its behavior mimics a uniaxial crystal. However, here, we assume a normal propagation of a TEM wave along the z direction. In this case, when the anisotropic nature is neglected, we

get the simplest closed-form solution. In particular, we start by finding the frequency shift law when the Fermi level of graphene is under a steplike change at time $t = t_0$, or, in other words, when plasma frequency is suddenly switched from ω_{p1} to ω_{p2} , as in Fig. 2. At this moment, the incident wave E_i is being divided into two [35,36]: one back-propagating in time (E_r) and one forward-propagating (E_t). For simplicity, we call them reflected and transmitted waves, respectively. The corresponding cases have the following forms in phasor notation:

$$E_i(t < t_0) = E_0 e^{j(\omega_i t - k_i z)}, \quad (7a)$$

$$E_r(t > t_0) = E_{r0} e^{j(\omega_r t - k_r z)}, \quad (7b)$$

$$E_t(t > t_0) = E_{t0} e^{j(\omega_t t - k_t z)}. \quad (7c)$$

Under any temporal discontinuity, the wave numbers of incident (subscript i) and scattered (subscript s) waves should be conserved [37], i.e., $k_i = k_s$. From this point, we obtain the following relation:

$$\frac{\omega_i}{c} \sqrt{\varepsilon_d - \frac{\omega_{p1}^2}{\omega_i^2}} = \frac{\omega_s}{c} \sqrt{\varepsilon_d - \frac{\omega_{p2}^2}{\omega_s^2}}. \quad (8)$$

Then, we derive the equation for the angular frequencies of scattered waves depending on the graphene Fermi level change from Eqs. (5) and (8):

$$\omega_s = \pm \sqrt{\omega_i^2 + \frac{\omega_{p2}^2 - \omega_{p1}^2}{\varepsilon_d}} = \pm \omega_i \sqrt{1 + \gamma \frac{\Delta \mu_F}{d \varepsilon_d \omega_i^2}}, \quad (9)$$

where $\Delta \mu_F$ is the difference between the initial and final Fermi level states and the positive sign of the scattered wave angular frequency indicates the transmitted wave ($s = t$), whereas the negative one indicates the reflected wave ($s = r$). Thus, the frequency shift for the metamaterial strongly depends on four parameters: the dielectric spacer permittivity and its thickness, initial frequency, and Fermi level change. Typical proportions between scattered and initial waves are shown in Fig. 3.

The conservation of the wave number, which arises from the continuity conditions, gives the values of frequencies for scattered waves but still does not give any information about their amplitudes. Meanwhile, Eq. (9) gives the restriction on the rapid negative change in the Fermi level, defining the maximal possible graphene Fermi level decrease as $\Delta \mu_F = -\omega_i^2 \varepsilon_d d / \gamma$. When this value is precisely reached, the frequency downshifting is not possible anymore, and it corresponds to the cutoff initial frequencies in Fig. 3 for specific graphene Fermi levels. Below them, the frequencies of the scattered waves become imaginary, which, after substitution, leads to functions exponentially varying in time. Proper understanding of different scenarios cannot be based only on analyzing Eq. (9) since the amplitudes of waves should also be taken into account. For this reason, to better understand whether the structure can support finite fields, after finding the solutions of the Maxwell equations in Sec. IV A, we implement the stability analysis in Sec. IV B.

In practical realizations, it is simpler to get a rapid carrier increase in graphene (i.e., an increase in the graphene Fermi level) than a decrease, for example, by laser

FIG. 3. Dependence of the frequency conversion on the difference in the Fermi level values of graphene. The following parameters are used: $d = 1 \mu\text{m}$ and $\varepsilon_d = 4$. Vertical dashed lines are cutoff frequencies: 0.205, 0.290, 0.355, and 0.410 THz.

pumping or applied voltage, so the rest of the paper is focused mainly on the rapid carrier increase.

IV. SCATTERING AMPLITUDES

A. Amplitude relations for scattered waves

The fields of scattered waves can be determined in several ways. The most common techniques in the electrodynamics of nonstationary media implement either the introduction of dimensionless timelike variables in Maxwell's or wave equations [38–40], integral equations [41–43], or eikonal equations [44]. Here, we use an approach which is commonly used for solving plasma physics tasks [26,35,45].

Let us start with Maxwell's equations. Assuming TEM waves propagating in the z direction (e^{-jkz} , where the k number is conserved during the modulation) with $\mathbf{E} = E \cdot \mathbf{a}_x$, $\mathbf{J} = J \cdot \mathbf{a}_x$, and $\mathbf{H} = H \cdot \mathbf{a}_y$, Maxwell's equations reduce to

$$jkE = \mu_0 \frac{\partial H}{\partial t}, \quad (10a)$$

$$jkH = \varepsilon_0 \varepsilon_d \frac{\partial E}{\partial t} + J. \quad (10b)$$

In the case of a sudden decrease in the graphene Fermi level, the total current J reduces similarly to the step function after the time jump. If the Fermi level is increased, new electrons appear with zero current [28]. In other words, after a sudden change in carriers in graphene at $t = 0$, current J obeys Drude's time-domain equation [26,27]:

$$\frac{\partial J}{\partial t} = \varepsilon_0 \omega_{p2}^2 E. \quad (11)$$

The set of equations (10) and (11) can be solved using the Laplace transform (see the Appendix). From there, we get the three modes described below.

a. Forward-propagating mode.

$$\frac{E_{t0}}{E_0} = \frac{1}{2} \left(1 + \frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right), \quad (12a)$$

$$\frac{H_{t0}}{H_0} = \frac{E_{t0}}{E_0} \left(\frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right), \quad (12b)$$

$$J_{t0} = -j \frac{\varepsilon_0 \gamma \mu_{F2}}{d\omega_s} E_{t0}. \quad (12c)$$

b. Backward-propagating mode.

$$\frac{E_{r0}}{E_0} = \frac{1}{2} \left(1 - \frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right), \quad (13a)$$

$$\frac{H_{r0}}{H_0} = -\frac{E_{r0}}{E_0} \left(\frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right), \quad (13b)$$

$$J_{r0} = -j \frac{\varepsilon_0 \gamma \mu_{F2}}{d\omega_s} E_{r0}. \quad (13c)$$

c. Free-streaming mode.

$$\frac{E_f}{E_0} = 0, \quad (14a)$$

$$\frac{H_f}{H_0} = \frac{\gamma \Delta\mu_F}{d\varepsilon_d\omega_s^2}, \quad (14b)$$

$$J_f = j \frac{\varepsilon_0 \gamma \Delta\mu_F (\varepsilon_d \omega_i^2 - \gamma \frac{\mu_{F1}}{d})}{d\omega_i \omega_s^2 \varepsilon_d} E_0. \quad (14c)$$

The obtained results can also be represented in the form of a temporal scattering parameter matrix, which is useful for further stability analysis. S parameters connect the field amplitudes before and after the temporal jump. In this formalism, we have

$$\begin{aligned} \begin{pmatrix} E_1^{t>0} \\ E_2^{t>0} \end{pmatrix} &= \begin{pmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{pmatrix} \begin{pmatrix} E_1^{t<0} \\ E_2^{t<0} \end{pmatrix} \\ &= \begin{pmatrix} \frac{1}{2} \left(1 + \frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right) & \frac{1}{2} \left(1 - \frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right) \\ \frac{1}{2} \left(1 - \frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right) & \frac{1}{2} \left(1 + \frac{1}{\sqrt{1 + \gamma \frac{\Delta\mu_F}{d\varepsilon_d\omega_i^2}}} \right) \end{pmatrix} \\ &\quad \times \begin{pmatrix} E_1^{t<0} \\ E_2^{t<0} \end{pmatrix} \\ &= \hat{S}_0 \begin{pmatrix} E_1^{t<0} \\ E_2^{t<0} \end{pmatrix}. \end{aligned} \quad (15)$$

These S parameters represent a virtual two-port network, where S_{11} and S_{22} represent temporal transmission and S_{12} and S_{21} are temporal reflections. As one can see, here, $S_{11} = S_{22}$, and $S_{12} = S_{21}$, but relations exist only in specific situations. Particularly, in the case when $\text{Re}(\sigma) \neq 0$, these relations do not hold [46].

Thus, any rapid change in the carrier concentration in graphene causes splitting of the initial waves into two prop-

FIG. 4. (a) Real and (b) imaginary parts of the eigenvalues depending on the parameter α .

agating waves in opposite directions, producing currents oscillating with new frequencies. The third mode lacks any electric field, but it has a nonzero dc current and static magnetic field components, which vary in space harmonically. Similar free-streaming modes have applications in free-electron lasers, in which relativistic plasma density waves are used as wigglers for producing coherent radiation [26,47]. One should note that these free-streaming mode fields tend to zero when the Fermi level of graphene varies continuously [48].

B. Stability analysis

Stability analysis is one of the essential things when dealing with temporally modulated systems since it examines whether a dynamically modulated material can support finite fields [49]. One of the common approaches is based on the eigenvalue problem analysis [50], playing a crucial role in exceptional point evaluation [51]. Here, we adapt an approach based on the time-transitioning state matrix, which was demonstrated recently by Koutserimpas and Fleury for a generalized medium under temporal modulation [46]. Let us

FIG. 5. Stability graphs for $\omega_i/2\pi = 0.5$ THz and $d = 1 \mu\text{m}$, $\varepsilon_d = 4$ for different values of $\Delta\mu_F$: (a) $\Delta\mu_F = -2.5$ meV, (b) $\Delta\mu_F = -0.5$ meV, (c) $\Delta\mu_F = 1$ meV, and (d) $\Delta\mu_F = 5$ meV. Blue corresponds to propagating modes, while yellow shows the amplified and evanescent ones.

define the transmission matrix $\mathbf{T}_n$ as

$$\mathbf{T}_n = \begin{pmatrix} e^{j\omega_n t} & 0 \\ 0 & e^{-j\omega_n t} \end{pmatrix}, \quad (16)$$

where $n = i, s$. During the abrupt change in the Fermi level in graphene, the wave number is conserved, in contrast to the frequency of modes. In this situation, ω_i and ω_s are connected as in Eq. (9). Thus, the time-transitioning state matrix is as follows:

$$\hat{H}(\tilde{\tau}) = \mathbf{T}_i(\tau) \hat{S}_0^{-1} \mathbf{T}_s(\tilde{\tau} - \tau) \hat{S}_0, \quad (17)$$

where $\tilde{\tau}$ is the time after a sudden change in the Fermi level ($\tilde{\tau} > \tau$). The corresponding eigenvalues are

$$\lambda_{\pm} = \alpha \pm \sqrt{\alpha^2 - 1}, \quad (18)$$

where $\alpha = \frac{1}{2} \text{tr}(\hat{H})$. The stability condition requires complex eigenvalues; thus, $|\alpha| < 1$. For $|\alpha| > 1$, the eigenvalues are real, representing amplified and evanescent fields, and when

$|\alpha| = 1$, the exceptional points occur. The typical behavior of eigenvalues depending on α is presented in Fig. 4.

After some calculations, the stability parameter α is

$$\alpha = \cos(\sqrt{\omega_i^2 + \beta} \delta\tau) \cos(\omega_i \tau) - \frac{2\omega_i^2 + \beta}{2\omega_i \sqrt{\omega_i^2 + \beta}} \sin(\sqrt{\omega_i^2 + \beta} \delta\tau) \sin(\omega_i \tau), \quad (19)$$

where $\beta = \gamma \frac{\Delta\mu_F}{d\varepsilon_d}$ and $\delta\tau = \tilde{\tau} - \tau$. The stability graphs are presented in Fig. 5. The initial wave propagates with the frequency ω_i , and then the Fermi level energy of graphene is suddenly changed. As we can see, with increasing values of $\Delta\mu_F$, areas of unstable solutions also grow.

With the help of this analysis it has also been shown that in such media the parity-time symmetry can be achieved [46]. If the modulation is periodic, the system also becomes a parametric amplifier or a perfect absorber [50].

FIG. 6. Harmonics generation efficiency depending on the frequency conversion.

C. Efficiency of harmonic generation

The frequency conversion efficiency in a rapidly switched graphene metamaterial depends on the Fermi level change implicitly. In fact, the amplitudes of scattered waves [Eqs. (14) and (13)] depend on the frequency conversion coefficient ω_s/ω_i (see the Appendix). To illustrate this, the frequency harmonics generation efficiency is shown in Fig. 6.

For example, the second harmonic generation efficiency is 75% and 25% for the transmitted and reflected waves, respectively, and the efficiency is 60% and 40% for the same waves generated at the fifth harmonic frequency. Although frequency harmonic generation has also been studied using standard nonlinear optics techniques [52], the present approach does not depend on the intensity (i.e., extreme fields are not required); however, the stability condition should be taken into account.

D. Time domain and spectral analysis

The effect of frequency conversion is convenient to describe in the time domain. We consider a Gaussian pulse in the form of a sinusoidal carrier modulated by a slowly varying envelope [53,54]:

$$E_0(t) \propto e^{j\omega_0 t} e^{-\frac{t^2}{2\tau_0^2}}, \quad (20)$$

where τ_0 is the effective width of the pulse, ω_0 is a carrier frequency, and the envelope has a long duration in comparison with the carrier period, i.e., its half-maximum width $\Delta\omega = \frac{1}{\tau_0} \ll \omega_0$. Its spectrum can be found via Fourier transform as

$$E_0(\omega) = \sqrt{2\pi\tau_0^2} e^{-\frac{\tau_0^2(\omega-\omega_0)^2}{2}}. \quad (21)$$

Combining Eqs. (12), (13), and (21), one may evaluate the spectra of scattered waves depending on the different Fermi level change values. The corresponding case is shown

FIG. 7. A Gaussian pulse conversion provided by temporally modulated graphene metamaterial: (a) spectrum upshifting by rapidly grown carrier density depending on the Fermi level variation and (b) Gaussian pulse effective width τ_0 for the calculated spectrum. The following parameters were used: $\varepsilon_d = 4$ and $d = 1 \mu\text{m}$.

in Fig. 7. In our calculation, we assume the input pulse has a central frequency of 0.5 THz, and the effective pulse width is $\tau_0 = 5$ ps.

As can be seen from Fig. 7(a), the amplitudes of the transmitted waves decrease with an increasing change in the Fermi level, while the reflected ones show the opposite behavior; also, the pulse carrier frequency ω_0 transforms by the same laws as Eq. (9). The spectral width decreases as the Fermi level of graphene increases, which can be confirmed by growing τ_0 as a function of $\Delta\mu_F$ in Fig. 7(b). The behavior of the temporal scattering of these pulses is shown in Fig. 8 as the envelopes of the incident and scattered waves.

It should be noticed that the pulse can also be suppressed by a rapid decrease in the Fermi level of graphene. However, the limitations mentioned in Secs. III and IV A do not allow it to decrease with the same freedom as in the case of frequency upshifting.

FIG. 8. The envelopes of the Gaussian pulse time-scattering problem depending on different $\Delta\mu_F$: (a) $\Delta\mu_F = 2$ meV, (b) $\Delta\mu_F = 4$ meV, (c) $\Delta\mu_F = 6$ meV, and (d) $\Delta\mu_F = 8$ meV.

V. FEASIBILITY OF THE TIME-VARYING METAMATERIAL CONCEPT

The results obtained here can be proven experimentally. One of the possible ways to fabricate a metamaterial based on graphene's multilayer structure is described in [55], which can be scaled to lower frequencies. Graphene can be grown with the chemical vapor deposition (CVD) technique and transferred onto the required substrate with polymethyl methacrylate (PMMA) as the carrier material [56]. Graphene Fermi level variation of such a metamaterial may be achieved in several ways. One of the common technologies implements an ion gel layer and external voltage [57,58]. In this case, the Fermi level varies as a function of the applied voltage V_{dc} as $\mu_F = \hbar v_F \sqrt{\frac{\pi C V_{dc}}{e}}$, where C is the gate capacitance and v_F is the Fermi velocity. Using electrical gating up to 30 GHz, the switching rate can be achieved [59]. This type of tunability allows achieving high Fermi levels and has already been used for time-varying metasurfaces [60]. However, adding gate electrodes to each layer might be a complicated or even impossible task for fabrication. In

contrast, optical control of the Fermi level in graphene might be a desirable solution, and no gates are required in this case. When an optical pump illuminates the metamaterial, the Fermi level in graphene changes as $\mu_F = \frac{12v_F^2 \hbar \tau_r}{137k_B T} \frac{I}{\Omega}$, where τ_r is the recombination time, Ω is the angular frequency of the pumping radiation, and I is its intensity [61]. This method allows achieving smaller values of the graphene Fermi levels, although the desired performance of the metamaterial can be reached by changing the spacer parameters. Another advantage of the optically driven tunability of graphene's properties is its high speed (on a scale of a few hundred femtoseconds) [62].

Experimental measurements of the structure might be performed by utilizing terahertz pump-probe spectroscopy. A similar experimental technique for time-varying metasurfaces was proposed in Ref. [3]. In the case of correlated measurements with pump-probe techniques, one can obtain the temporal wave forms from a rapidly switched multilayered graphene-based medium. However, the finite sizes of the metamaterial cause multiple reflections at the interfaces, which should be taken into account.

The short-term prospects for the proposed topic of temporally modulated multilayered graphene metamaterials may be aimed at the propagation of TE and TM modes in such a metamaterial [30,31]. In the case of TM modes, the hyperbolic behavior of the photonic band structure for such types of metamaterials may open new horizons for advanced methods of electromagnetic field manipulation [63]. Moreover, adding time modulation to multilayer structures may also open new possibilities for light-trapping devices [64] and other integrated photonic systems [65].

Furthermore, the suggested approach can be extended to the case of the inhomogeneity of graphene. Real CVD-grown graphene generally takes the form of flakes, which results in localization of the carriers inside the flake. Conductivity in such a system can be described by the Drude-Smith formalism [66] instead of the simple Drude model used here. That approach can mimic the carrier conductivity behavior observed in graphene nanoflakes.

VI. SUMMARY AND CONCLUSIONS

In this paper, we have discussed a temporally modulated metamaterial based on a graphene multilayered structure. We have revealed that after a rapid change in the Fermi level in graphene on the scale of a few meV the incident wave splits into two waves with the same shifted frequencies but different amplitudes. The frequency of time-scattered waves is proportional to the graphene Fermi level change and independent of its initial value. Frequency shifting of scattered waves depends on the sign of the Fermi level change. Negative values of the Fermi level change lead to downshifting of the frequency. However, a further decrease in the Fermi level may lead to the decay of the scattered waves. Increasing the graphene Fermi level results in frequency upshifting. In this case, the amplitude of the transmitted wave decreases, while the amplitude of the reflected wave increases with a greater Fermi level change. The proposal for a possible experimental time modulation scheme verified by terahertz pump-probe spectroscopy was outlined. The demonstrated frequency conversion and pulse broadening under a rapid graphene Fermi level change by a few meV open new paths for the development of terahertz harmonic generators operating without high-intensity pumping, as well as magnet-free nonreciprocal devices.

APPENDIX: DERIVATION OF THE SOLUTION OF MAXWELL'S EQUATIONS USING THE LAPLACE TRANSFORM

This set of equations can be solved by implementing the commonly used Laplace transform:

$$\mathcal{L}[F(t)] = \int_{0+}^{\infty} F(t)e^{-st} dt = \tilde{F}(s), \quad (\text{A1a})$$

$$\mathcal{L}\left[\frac{dF(t)}{dt}\right] = s\tilde{F}(s) - F(0^+). \quad (\text{A1b})$$

After that, the set of equations (10) and (11) transforms into the set of linear algebraic equations

$$jk\tilde{E} = \mu_0[s\tilde{H} - H(0)], \quad (\text{A2a})$$

$$jk\tilde{H} = \varepsilon_0\varepsilon_d[s\tilde{E} - E(0)] + \tilde{J}, \quad (\text{A2b})$$

$$s\tilde{J} - J(0) = \varepsilon_0\omega_{p2}^2\tilde{E}, \quad (\text{A2c})$$

where $E(0) = E_0$, $H(0) = H_0$, and $J(0) = J_0$. The solution can be found using standard linear algebra techniques as follows:

$$\tilde{H} = -\frac{H_0\varepsilon_0\mu_0\omega_{p2}^2 + H_0\varepsilon_0\varepsilon_d\mu_0s^2 + jE_0\varepsilon_0\varepsilon_dks - J_0kj}{s(k^2 + \varepsilon_0\mu_0\omega_{p2}^2 + \varepsilon_0\varepsilon_d\mu_0s^2)}, \quad (\text{A3a})$$

$$\tilde{E} = \frac{\mu_0(E_0\varepsilon_0\varepsilon_d s - J_0 + H_0kj)}{k^2 + \varepsilon_0\mu_0\omega_{p2}^2 + \varepsilon_0\varepsilon_d\mu_0s^2}, \quad (\text{A3b})$$

$$\tilde{J} = \frac{E_0\varepsilon_d\mu_0\varepsilon_0^2\omega_{p2}^2s + jH_0\mu_0\varepsilon_0k\omega_{p2}^2 + J_0\varepsilon_d\mu_0\varepsilon_0s^2 + J_0k^2}{s(k^2 + \varepsilon_0\mu_0\omega_{p2}^2 + \varepsilon_0\varepsilon_d\mu_0s^2)}. \quad (\text{A3c})$$

To find the amplitudes of scattered fields E , we use the following initial conditions:

$$H_0 = \sqrt{\frac{\varepsilon_0}{\mu_0}}\sqrt{\varepsilon_1}E_0, \quad (\text{A4a})$$

$$J_0 = -j\omega_{p1}^2\frac{\varepsilon_0}{\omega_i}E_0. \quad (\text{A4b})$$

The calculation of fields in normal space was performed using the inverse Laplace transform $\mathcal{L}^{-1}\tilde{F}(s) = F(t)$. There are several ways to implement the inverse Laplace transform. The first one is to use the table of inverse Laplace transformations. A more general technique uses finding the residues and contour integration of the Bromwich integral [67]. Both of them are mostly included in modern mathematical software (for instance, MATLAB). Calculating the inverse transformation by any convenient technique, one gets the following results for the fields at $t > 0$:

$$E(t) = \frac{E_0}{2} \left(e^{j\omega_s t} + e^{-j\omega_s t} + \frac{\omega_i\sqrt{\varepsilon_d}(e^{j\omega_s t} - e^{-j\omega_s t})}{\sqrt{\varepsilon_d\omega_i^2 - \omega_{p1}^2 + \omega_{p2}^2}} \right) = \frac{E_0}{2} \left[\left(1 + \frac{\omega_i}{\omega_s}\right)e^{j\omega_s t} + \left(1 - \frac{\omega_i}{\omega_s}\right)e^{-j\omega_s t} \right], \quad (\text{A5a})$$

$$H(t) = \frac{H_0}{2} \left[(e^{j\omega_s t} + e^{-j\omega_s t}) \frac{\omega^2\varepsilon_d}{\omega^2\varepsilon_1 + \omega_{p2}^2} + \frac{\omega_i\sqrt{\varepsilon_d}(e^{j\omega_s t} - e^{-j\omega_s t})}{\sqrt{\varepsilon_d\omega_i^2 - \omega_{p1}^2 + \omega_{p2}^2}} \right] = \frac{H_0}{2} \frac{\omega_i}{\omega_s} \left[\left(1 + \frac{\omega_i}{\omega_s}\right)e^{j\omega_s t} - \left(1 - \frac{\omega_i}{\omega_s}\right)e^{-j\omega_s t} \right], \quad (\text{A5b})$$

$$H_f = H_0 \frac{\omega_{p2}^2 - \omega_{p1}^2}{\omega_i^2\varepsilon_1 + \omega_{p2}^2}, \quad (\text{A5c})$$

$$J_f = \frac{\varepsilon_1 E_0 j \omega \varepsilon_0 (\omega_{p2}^2 - \omega_{p1}^2)}{\varepsilon_1 \omega^2 + \omega_{p2}^2} = j \frac{\varepsilon_0 E_0 (\varepsilon_d \omega^2 - \omega_{p1}^2) (\omega_{p2}^2 - \omega_{p1}^2)}{\omega_i \omega_s^2 \varepsilon_d}, \quad (\text{A5d})$$

$$J(t) = -j \frac{E_0}{2} \left[\frac{\varepsilon_0 \sqrt{\varepsilon_d} \omega_{p2}^2 (e^{j\omega_s t} - e^{-j\omega_s t})}{\sqrt{(\omega_i^2 \varepsilon_1 + \omega_{p2}^2)}} + \frac{\varepsilon_0 \omega_{p2}^2 (\varepsilon_1 \omega_i^2 + \omega_{p1}^2) (e^{j\omega_s t} + e^{-j\omega_s t})}{\omega_i (\varepsilon_1 \omega_i^2 + \omega_{p2}^2)} \right] = -j \frac{\varepsilon_0 \omega_{p2}^2}{\omega_s} E(t). \quad (\text{A5e})$$

After substituting the initial dielectric permittivity $\varepsilon_1 = \varepsilon_d - \frac{\omega_{p1}^2}{\omega_i^2}$ and Eqs. (7)–(9), we get the formulas in the main text.

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