Homogenization and Nonlinearity Enhancement of 2D Graphene-Based Metasurfaces

Jian Wei You and Nicolae C. Panoiu

Department of Electronic and Electrical Engineering, University College London, London, WC1E7JE, UK.

j.you@ucl.ac.uk and n.panoiu@ucl.ac.uk

Abstract: A general homogenization technique is developed to study the linear and nonlinear properties of 2D graphene-based metasurfaces. The results show the effective nonlinear susceptibility of graphene metasurfaces can be enhanced by two orders of magnitude.

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1. Introduction

We introduce a general linear and nonlinear homogenization method for 2D graphene-based metamaterials to overcome some shortcomings of existing linear homogenization methods. Based on our novel method, a graphene cruciform metasurface is studied and the results show its effective third-order susceptibility can be enhanced by more than two orders of magnitude as compared to that of a homogeneous graphene sheet.

As is well known, homogenization methods are key tools to study novel properties of metamaterials [1, 2]. So far, the scattering-parameter approach and the field-averaging procedure are the two most commonly used homogenization methods [3]. However, they have been mostly applied to simple 3D metamaterials (linear, dispersive, and isotropic case), and almost no efforts have been devoted to the nonlinear effective properties of 3D metamaterials [4]. As for metasurfaces, relevant work is even more scarcely found. One of the main reasons is that the high-order susceptibility of 2D materials exhibits optical anisotropy, which nontrivially challenges the existing linear homogenization methods. To overcome these challenges, an auxiliary physical quantity is introduced here to develop a general linear and nonlinear homogenization method for 2D graphene-based metasurfaces.

2. Linear Homogenization of Graphene Metasurfaces

In a widely used linear field-average homogenization method [3], the effective permittivity is evaluated as $\varepsilon_i^{av} = D_i^{av}/E_i^{av}$. Apparently, this method is only applicable to some particular anisotropic materials, whose permittivity tensor is diagonal. In order to extend the field-average method to a more general case, we introduce here a new auxiliary quantity, d_{ij}, defined as $d_{ij} = \varepsilon_{ij}E_j$. Based on this auxiliary quantity, the constitutive relation of a general anisotropic material is expressed as $D_i = \sum_j d_{ij}$. If we define the average value of each auxiliary quantity component as $d_{ij}^{av}(\omega) = \int_V \varepsilon_{ij}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)d\mathbf{r}/V$, where V is the volume of the unit cell, the corresponding component of effective permittivity tensor can be evaluated as $\varepsilon_{ij}^{av}(\omega) = d_{ij}^{av}(\omega)/E_j^{av}(\omega)$.

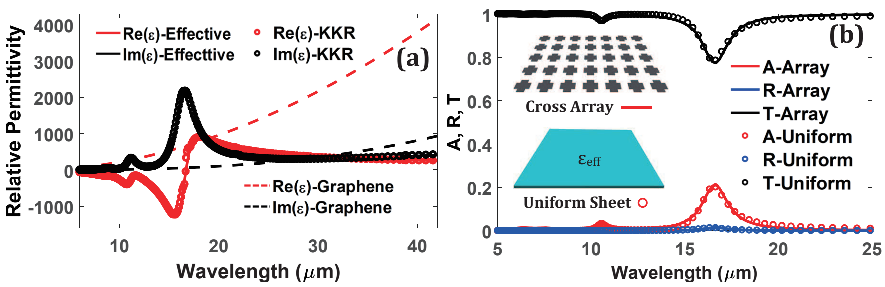

Fig. 1. Validation of linear homogenization: (a) Comparison of intrinsic graphene permittivity, the effective metasurface permittivity, and the effective permittivity constructed from the Kramers-Kronig relation. (b) Comparison of linear response of the graphene metasurface and homogenized one.

In order to validate our proposed linear homogenization approach, a graphene cruciform metamaterial has been studied by the FDTD method, the relevant results being presented in Fig. 1. The comparison of intrinsic and effective permittivities shown in Fig. 1(a) clearly demonstrates that the optical properties of the graphene array differ significantly from those of the homogeneous graphene sheet, chiefly due to the presence of plasmon resonances. Moreover, the effective permittivity satisfies the well-known Kramers-Kronig relation, which further proves the accuracy of the proposed linear homogenization method. In Fig. 1(b), the comparison between the linear response of the structured and homogenized metasurfaces proves that a complex graphene metasurface can be replaced by a homogenized sheet with specific optical constants.

3. Nonlinear Homogenization of Graphene Metamaterials

Similarly to the linear case, we introduce an auxiliary quantity q_{ijik}, defined in third-harmonic case as $q_{ijkl} = \chi^{(3)}_{ijkl}E_jE_kE_l$. Based on the field-average method, we have $q^{av}_{ijkl}(\omega) = \int_V \chi^{(3)}_{ijkl}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)E_k(\mathbf{r},\omega)E_l(r,\omega)d\mathbf{r}/V$. To generate the same THG intensity from an effective uniform sheet, we assume each component of q^{eff}_{ijkl} in a homogeneous uniform sheet is equal to that of q^{av}_{ijkl} in the graphene cruciform metasurface. Based on this assumption, we have $\chi^{(3),eff}_{ijkl} = q^{av}_{ijkl}/(E^{eff}_jE^{eff}_kE^{eff}_l)$. Using an in-house developed GS-FDTD code [5], we have determined the nonlinear optical response of the structured and homogenized metasurfaces, the main results being summarized in Fig. 2. Inspecting the plots in Fig. 2(a), we find that, at the resonance wavelength of 5.4 µm, the effective $\chi^{(3)}$ is more than two orders of magnitude larger than the intrinsic third-order susceptibility of graphene. Moreover, we have compared the nonlinear optical response of the structured and homogenized graphene metasurfaces and the results are summarized in Fig. 2(b). The good agreement proves the effectiveness and accuracy of our proposed nonlinear homogenization method.

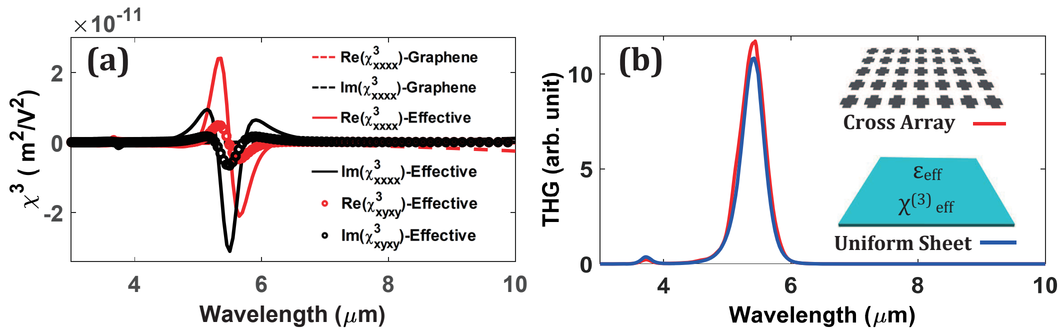

Fig. 2. Validation of the nonlinear homogenization method: (a) Comparison of intrinsic and effective third-order susceptibilities. (b) Comparison of third-harmonic generated by the two metasurfaces.

To conclude, we prove that the third-order susceptibility of homogenous graphene sheet can be enhanced more than two order of magnitude by designing a graphene metasurface properly. This remarkable enhancement can find important applications to active nanodevices, as it offers a new avenue to design metasurfaces with extremely large nonlinear susceptibilities at desirable frequencies.

References

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