Previously challenging spectral blue-shifting of plasmonic metasurfaces is enabled by overlaying them with single-layer graphene, paving the way for next-generation tunable infrared optoelectronic devices.

Metamaterials are made of engineered elements that, similarly to natural atoms and molecules, are capable of interacting especially strongly with light of certain frequencies: a property called resonant behavior. Optical metamaterials and metasurfaces have great potential for photonic and optoelectronic applications due to their unusual electromagnetic properties, such as optical magnetism and negative refraction. They are typically comprised of resonant plasmonic elements with a spectrally narrow response, i.e. interacting only with light inside a narrow frequency range. The ability to actively control the optical response of metamaterials, both by red-shifting and blue-shifting their resonant frequencies, has potential to expand the range of their applications even further. A variety of approaches to achieve such tunability have emerged recently, with most techniques relying on integrating metamaterials with various optically active materials such as semiconductors, liquid crystals and nonlinear media. However, most of these approaches result in the red-shifting of the resonances: blue-shifting has been a challenge so far.

The concept of frequency tuning is best understood by representing a resonant metasurface element as an LC circuit with resonance frequency \( \omega = 1/\sqrt{LC} \). The addition of optically active materials to the metasurface typically increases the capacitance \( C \) due to field expulsion and decreases the inductance \( L \) due to image currents. However, with most conventional materials, a change in \( C \) usually dominates a change in \( L \), making the blue-shifting more challenging to achieve than the red-shifting. Recently, we proposed a novel approach to blue-shifting metamaterials’ resonances in the mid-IR part of the spectrum by enveloping them in a zero-volume low-loss plasmonic material: single-layer graphene (SLG). This is a one-atom-layer-thick carbon allotrope (structural form) that has a strongly anisotropic optical response: it has high in-plane conductivity and essentially vanishing out-of-plane conductivity that does not affect the capacitance. However, as we showed both theoretically and experimentally, it modifies the inductance, thus giving rise to blue-shifting. We also demonstrated that by measuring the spectral shift and the broadening of the metamaterial resonance due to the presence of SLG, it is possible to extract electronic properties such as Fermi energy and carrier-scattering rate.

To illustrate the uniqueness of the mid-IR spectral range for graphene applications, we plotted the ratio of the reactive to resistive conductivities (given by the imaginary, \( \text{Im} \), and real, \( \text{Re} \), parts of the surface conductivity \( \sigma \), respectively) of moderately doped graphene for wavelengths of light ranging from 1 to 100\( \mu \)m: see Figure 1(a). Note that this conductivity is quite different from the DC conductivity and it originates from both intra- and inter-band high-frequency transitions. Clearly, mid-IR is the only spectral window where \( \text{Re}(\sigma) \ll \text{Im}(\sigma) \), i.e., where graphene acts as a low-loss material. However, because of the weak response of graphene in this region, most earlier research on its optical properties focused on the low-frequency (terahertz)
or high-frequency (visible) ranges and largely overlooked the mid-IR window. While others showed that the graphene’s resistive response in the visible and terahertz spectral ranges can be profitably used for amplitude modulation, these spectral ranges have limited appeal for low-loss spectral tuning of metamaterials.

To demonstrate the possibility of inductive tuning in the mid-IR due to the plasmonic response of SLG, we used a metasurface that supports multiple resonances interacting with each other (a Fano-resonant metasurface). This is exemplified by a two-dimensional periodic array of $\pi$-shaped plasmonic metamolecules comprised of three antennas: see inset of Figure 2(b). The structure supports two resonances with disparate lifetimes: a low-Q (≈3) dipolar and a high-Q (≈9) quadrupolar resonances, where Q is the quality factor that characterizes the ability of the structure to trap photons. The interference between the two resonances results in a spectrally sharp Fano feature. Figure 1(b) predicts how the spectral position of both resonances blue-shifts when a moderately doped SLG is transferred onto the metasurface. The magnitude of the blue-shift increases monotonically with the Fermi energy of SLG. Due to the larger field enhancement, the shift of the quadrupolar resonance is more pronounced compared to that of the dipolar resonance.

We fabricated two samples (each comprised of $30 \times 30$ unit cells) with scaled geometric dimensions on a quartz substrate: see Figure 2. We measured the reflectivity before and after the dry transfer of graphene using a Fourier transform infrared spectrometer: see Figure 2(a). Figure 2 (b) and (c) shows the intensity of the Raman-active G-band and the Raman spectra obtained at different locations on graphene, which confirms the presence of a high-quality SLG on our samples. We determined the spectral positions and linewidths of the dipolar and quadrupolar resonances before and after the graphene transfer by fitting the experimental reflectivity spectra. As anticipated, the quadrupolar resonance was spectrally blue-shifted by a large amount ($\approx 30\text{cm}^{-1}$) for both samples. Further, the linewidth for the quadrupolar resonance stemming from the additional resistive loss in graphene changed noticeably. This data also allowed us to extract the Fermi energy and the carrier scattering rate, which we found to be 0.2eV and 269cm$^{-1}$, respectively. The Fermi energy obtained from this spectroscopic analysis agreed well with that obtained using Raman spectroscopy. An unintentional chemical doping of graphene during the transfer process caused the non-zero value of the Fermi level.

In summary, we have proposed that by overlaying a one-atom-thick graphene layer on multi-resonant metasurfaces, we can achieve an uncommon blue-shift tunability due to the inductive coupling between the graphene layer and the underlying metasurface. We experimentally demonstrated that the transfer of chemically doped graphene onto the metasurface results in a blue-shift of the resonances as high as $30\text{cm}^{-1}$. The potential of controlling the optical response of the multi-resonant metasurfaces, combined with the ability to dynamically control its Fermi energy, paves the way for the next-generation of tunable infrared optoelectronic devices. In the future we aim to dynamically tune the metamaterial resonances using a method called electrostatic gating, which enables rapid real-time control of a metamaterial’s response.

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References