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Mechanically reconfigurable architected graphene for tunable plasmonic resonances

Pilgyu Kang^{1,2}, Kyoung-Ho Kim³, Hong-Gyu Park^{3,4} and SungWoo Nam¹

Abstract

Graphene nanostructures with complex geometries have been widely explored for plasmonic applications, as their plasmonic resonances exhibit high spatial confinement and gate tunability. However, edge effects in graphene and the narrow range over which plasmonic resonances can be tuned have limited the use of graphene in optical and optoelectronic applications. Here we present a novel approach to achieve mechanically reconfigurable and strongly resonant plasmonic structures based on crumpled graphene. Our calculations show that mechanical reconfiguration of crumpled graphene structures enables broad spectral tunability for plasmonic resonances from mid- to near-infrared, acting as a new tuning knob combined with conventional electrostatic gating. Furthermore, a continuous sheet of crumpled graphene shows strong confinement of plasmons, with a high near-field intensity enhancement of $\sim 1 \times 10^4$. Finally, decay rates for a dipole emitter are significantly enhanced in the proximity of finite-area biaxially crumpled graphene flakes. Our findings indicate that crumpled graphene provides a platform to engineer graphene-based plasmonics through broadband manipulation of strong plasmonic resonances.

Introduction

Graphene is a promising plasmonic material with superior properties that distinguish it from conventional noble metals^{1,2}, including relatively low optical loss, high spatial confinement, and tunable plasmonic resonances. Graphene-based plasmonic devices have shown potential in broad applications, including optical biosensing³, optical communications⁴, photodetectors⁵, and plasmonic metamaterials⁶. In particular, desired plasmonic resonances are easily excited in lithographically patterned graphene structures^{4,6–14} using a far-field coupling method. However, such structural patterns in graphene are not reconfigurable, thus fixing their resonance

wavelengths. Although one can tune the resonance wavelength of graphene through electrical gating^{7,11,15}, it is challenging to achieve broadband tunability across the near- to mid-infrared wavelength ranges. While graphene grating structures have been proposed for on–off switching and plasmonic modulation^{16–19}, post-fabrication reconfiguration of plasmonic structures has not yet been realized.

It has recently been demonstrated that mechanical crumpling of graphene can be used to create different surface structures while maintaining reversibility^{20–22}. However, the potential of crumpled graphene structures for reconfigurable plasmonic materials has not been explored. In this work, we report on the use of mechanically reconfigurable crumpled graphene structures to realize strong plasmonic resonances with broadband tunability. We introduce a resonant electrical inductor–capacitor (*LC*)-circuit model to describe the characteristics of plasmonic resonances in the crumpled graphene. By varying the geometry of the crumpled

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graphene structures, we show a tunable spectral range for plasmonic resonances that is fivefold broader than that achieved by conventional electrical gating. Our results show that mechanical reconfiguration of crumpled graphene serves as a new way to manipulate strong plasmonic resonances over a broad spectral range.

Materials and methods

Numerical simulations

We performed full-wave simulations using the finite element method (COMSOL Multiphysics, wave optics module) to investigate the plasmonic resonances of crumpled graphene structures. We modeled graphene as a two-dimensional surface current tangential to the crumpled structure: $\mathbf{J}_s = \sigma \mathbf{E}_t$, where $\mathbf{J}_s$ is the surface current, σ is the optical conductivity of graphene, and $\mathbf{E}_t$ is the tangential component of the electric field to the graphene surface^{23,24}. We used a mobility value of 10,000 cm²/(V·s), which is achievable for crumpled graphene based on the reported range of mobilities (μ) for flat graphene¹⁵ from ~1000 cm²/(V·s) for chemical vapor deposition grown graphene to 230,000 cm²/(V·s) for suspended exfoliated graphene. Moreover, the mobility of wrinkled graphene was shown to be similar to that of flat graphene^{25,26}. For the optical absorption/extinction spectra of uniaxially and biaxially crumpled graphene in Figs. 1–3, we performed absorption/transmission calculations with periodic boundary conditions imposed on the respective crumpling directions. The optical extinction spectra were obtained by calculating $1 - T/T_0$, where T and T_0 are the transmittance values with and without crumpled graphene, respectively. To investigate substrate-induced effects on plasmonic resonances in Fig. 3a, a silicon oxide (SiO₂) substrate²⁷ was used as a model material. In Fig. 4a, we obtained the extinction cross-section for a normal-incident electromagnetic field by integrating over the scattered light power and optical loss in graphene. In Fig. 4b, c, we calculated the enhancement of the radiative decay rates by taking the ratio of the radiated light power from a dipole emitter with a crumpled graphene flake to that without the crumpled graphene flake. In Fig. 4, we applied the perfectly matched layer for all boundaries of the calculation domain to reduce reflections from the boundaries.

Optical conductivity of graphene

We calculated the optical conductivity of graphene (σ) for varying Fermi energy (E_F) and carrier mobility (μ) values of graphene at a constant temperature of 300 K using a random phase approximation method^{23,24,28,29}. The optical conductivity is expressed by $\sigma(\omega) = \sigma_{\text{intra}}(\omega) + \sigma_{\text{inter}}(\omega)$, where $\sigma_{\text{intra}}(\omega)$ and $\sigma_{\text{inter}}(\omega)$ are the intraband and interband conductivities, respectively²³. They are expressed as below, where ω is the frequency of the

incident light, e is the charge of an electron, τ is the Drude relaxation rate, T is the temperature, k_B is the Boltzmann constant, and $\hbar$ is the reduced Planck constant. E_F is the Fermi energy, determined by $E_F = \hbar v_f(\pi n)^{1/2}$, where v_f is the Fermi velocity of electrons (~10⁶ m/s) and n is the carrier density of graphene. The Drude relaxation rate τ is determined by $\tau = \mu E_F / e v_f^2$, where μ is the carrier mobility of graphene.

$$\sigma_{\text{intra}}(\omega) = \frac{2e^2\omega_T}{\pi\hbar} \frac{i}{\omega + i\tau^{-1}} \log \left[2 \cosh \left(\frac{\omega_F}{2\omega_T} \right) \right] \quad (1)$$

$$\sigma_{\text{inter}}(\omega) = \frac{e^2}{4\hbar} \left[H \left(\frac{\omega}{2} \right) + i \frac{2\omega}{\pi} \int_0^\infty \frac{H \left(\frac{\omega'}{2} \right) - H \left(\frac{\omega}{2} \right)}{\omega^2 - \omega'^2} d\omega' \right] \quad (2)$$

$$H(\omega) = \frac{\sinh(\omega/\omega_T)}{\cosh(\omega_F/\omega_T) + \cosh(\omega/\omega_T)} \quad (3)$$

where $\omega_F = E_F/\hbar$ and $\omega_T = k_B T/\hbar$.

Results and discussion

Plasmonic resonances of crumpled graphene structures

To study the plasmonic resonances of the crumpled graphene structures, we modeled crumpled graphene as a continuous sinusoidal structure with geometrical parameters, including crumple wavelength (λ_c) and crumple height (h) (upper inset of Fig. 1a). First, we performed full-wave optical simulations (see Materials and methods) to investigate the far-field spectral response (i.e., optical absorption spectra) of free-standing uniaxially crumpled graphene (Fig. 1a). The incident electric field ($\vec{E}_{\text{ext}}$) is parallel and perpendicular to the crumpling direction ($\vec{\lambda}_c$) ($\vec{E}_{\text{ext}} \parallel \vec{\lambda}_c$ and $\vec{E}_{\text{ext}} \perp \vec{\lambda}_c$, respectively). Our simulation result including the consideration of strain effects on crumpled graphene^{30–32} was very similar to the result obtained without consideration of strain effects (Fig. S1). In the optical absorption spectrum, we observed multiple resonance peaks, including the first- and higher-order (second- and third-order) plasmonic modes (lower insets in Fig. 1a) for $\vec{E}_{\text{ext}} \parallel \vec{\lambda}_c$, whereas no resonance peak was observed for $\vec{E}_{\text{ext}} \perp \vec{\lambda}_c$. These far-field resonance peaks for $\vec{E}_{\text{ext}} \parallel \vec{\lambda}_c$ are attributed to resonant graphene plasmons confined in the crumpled graphene structures^{9,16,33}. The most efficient coupling of the incident light to the graphene plasmon is observed in the peak of the maximum optical absorption (Fig. 1a). In subsequent investigations, we focus on this strongest plasmonic resonance.

We further investigated plasmonic resonances in crumpled graphene for various λ_c and h . First, we observed the optical absorption and extinction spectra for different aspect ratios of crumple height to wavelength (h/λ_c) at a constant λ_c of 250 nm (Fig. 1b and Fig. S2). The plasmonic resonance wavelength (λ_{res}) redshifted from ~4.5 to ~11.3 μm with increasing h/λ_c from 0.25 to 2.0. The optical absorption and extinction (σ_{abs} and σ_{ext}) also

Fig. 1 Mechanically reconfigurable plasmonic resonances of crumpled graphene structures. **a** Optical absorption spectra of a crumpled graphene structure with $\lambda_c = 250$ nm and $h/\lambda_c = 1$, where λ_c is the crumpled wavelength and h is the crumple height, for $\vec{E}_{ext} \parallel \vec{\lambda}_c$ (black) and $\vec{E}_{ext} \perp \vec{\lambda}_c$ (red), where $\vec{E}_{ext}$ is the incident electric field and $\vec{\lambda}_c$ is the crumpling direction vector. Fermi energy E_F and carrier mobility μ are set to 0.64 eV and 10,000 $\text{cm}^2/(\text{V}\cdot\text{s})$, respectively. The upper inset shows a schematic illustration for the uniaxially crumpled graphene structure. The lower insets show field intensity distributions of the first-, second-, and third-order modes at wavelengths of 6.94, 4.58, and 3.74 μm , respectively. **b** Optical absorption spectra for the uniaxially crumpled graphene structures with varying h/λ_c from 0.25 to 2.0 at $\lambda_c = 250$ nm, $E_F = 0.64$ eV, and $\mu = 10,000$ $\text{cm}^2/(\text{V}\cdot\text{s})$. **c** Near-field distributions normalized by the incident field (E_0) in the uniaxially crumpled graphene structures at the plasmonic resonance wavelengths λ_{res} denoted in **b**

increased with higher h/λ_c from 0.25 to 1.0 but showed no significant change from $h/\lambda_c = 1.0$ to 2.0 (Fig. S2a, c, e). Next, we varied λ_c from 50 nm to 1 μm with a fixed aspect ratio ($h/\lambda_c = 1$) and observed redshifts for λ_{res} from ~ 3.2 to ~ 13.8 μm and an increase in σ_{abs} and σ_{ext} from ~ 0.12 to ~ 0.41 and from ~ 0.14 to ~ 0.50 , respectively (Fig. S2b, d, f). Moreover, near-field distributions for graphene plasmons varied with different h/λ_c values (Fig. 1c). An enhanced near-field intensity near the apex and valley regions was manifested with increasing h/λ_c . We observed a maximum near-field intensity enhancement of $\sim 40^2$ in the apex and valley regions of crumpled graphene structures at $h/\lambda_c = 1$. Taken together, these results demonstrated the tunability of both far- and near-field plasmonic resonances by the reconfiguration of crumpled graphene structures.

Analytical description of the plasmonic resonance in crumpled graphene structures

To describe the plasmonic resonance in crumpled graphene structures, we introduced a resonant LC -circuit model based on the electric field and current distributions

(Fig. 2a). An effective inductance L_{eff} and effective capacitance C_{eff} are used to account for the characteristics of the current and charge induced by the graphene plasmons, respectively^{34,35}. By using the crumpled structure parameters, including λ_c and h , as well as the electronic properties of graphene, we derived L_{eff} and C_{eff} as follows:

$$L_{eff} = \frac{\pi \hbar^2}{e^2 E_F} \lambda_c, C_{eff} = \frac{\epsilon_0}{2\alpha} \ln[1 + (1 + e^{-\alpha})] \quad (4)$$

where $\hbar$ is the reduced Planck constant, e is the elementary charge, ϵ_0 is the permittivity in vacuum, and $\alpha = \pi - 2\arctan(2h/\lambda_c)$ is the curvature factor for the crumpled structures. By using L_{eff} and C_{eff} , we derived the plasmonic resonance wavelength (λ_{res}) of the crumpled graphene expressed as $\lambda_{res} = 2\pi c(L_{eff}C_{eff})^{1/2}$ (see SI for detailed derivation).

To validate our LC -circuit model, we compared the value for λ_{res} predicted by the LC -circuit model with that predicted by numerical simulations. With varying λ_c from 50 nm to 1 μm and h/λ_c from 0.5 to 2, the λ_{res} obtained by the full-wave simulations (dots) agreed with that of our

Fig. 2 Geometry dependence of the plasmonic resonances in uniaxially crumpled graphene structures. **a** Normalized near-field distribution and electric field vectors. The right panel shows the normalized induced current distribution for the region indicated by a dotted square in the left panel. The inset of the right panel shows an LC-circuit model that describes the characteristics of resonant plasmons in the crumpled graphene structures. **b** Plasmonic resonance wavelengths λ_{res} for crumpled graphene structures (dots) with varying λ_c from 50 nm to 1 μm at constant h/λ_c . The h/λ_c varies from 0.5 to 2. The curves calculated by the LC-circuit model (solid curves) and the conventional model (a dotted curve) are plotted together. **c** The λ_{res} of uniaxially crumpled graphene structures (dots) with varying E_F from 0.24 to 0.84 eV at $\lambda_c = 100$ nm, $h/\lambda_c = 1$, and $\mu = 10,000$ $\text{cm}^2/(\text{V.s})$. The λ_{res} was also calculated as a function of E_F using the LC-circuit model (solid curve). The inset shows the optical absorption spectra of uniaxially crumpled graphene structures for various μ from 1000 to 10,000 $\text{cm}^2/(\text{V.s})$ at $E_F = 0.64$ eV, $\lambda_c = 100$ nm and $h/\lambda_c = 1$

LC-circuit model (solid curves) (Fig. 2b). In contrast, conventional analytical models account for only crumple wavelengths of a periodically modulated graphene sheet^{16–19} and cannot predict changes in λ_{res} in response

to varying h/λ_c (dashed curve in Fig. 2b). The C_{eff} changes with varying h/λ_c (i.e., change of α), leading to a shift in λ_{res} . This implies that controlling the charge interactions on confronting graphene surfaces by varying h/λ_c enables

the tunability of λ_{res} across the near- to mid-infrared wavelength ranges. The mechanically reconfigurable crumpled graphene structures are thus an excellent platform for broadband tunability of plasmonic resonances.

To compare the wavelength tunability of the plasmonic resonances by mechanically reconfigurable crumpled graphene structures to that achievable by conventional electrical gating, we studied the electrical tunability of the plasmonic resonances in uniaxially crumpled graphene by varying the electronic properties of graphene such as the Fermi energy E_F and carrier mobility μ (Fig. 2c). In our simulation, λ_{res} blueshifted from ~ 7.6 to ~ 3.9 μm with increasing E_F from 0.24 to 0.84 eV (Fig. 2c), showing the relation $\lambda_{\text{res}} \propto E_F^{-1/2}$ as predicted by our LC-circuit model (a solid curve in Fig. 2c). Additionally, the σ_{abs} increased with higher μ (inset of Fig. 2c). Most notably, the tunable range of the plasmonic resonance wavelengths ($\Delta\lambda_{\text{res}}$) for varying E_F was ~ 3.7 μm , while $\Delta\lambda_{\text{res}}$ for varying λ_c was ~ 20.1 μm ($\Delta\lambda_{\text{res}} = \lambda_{\text{res,max}} - \lambda_{\text{res,min}}$ in Fig. 2b). These results show broad spectral tunability from the mid- to near-infrared by the mechanical reconfiguration of crumpled graphene structures, which allows for a new tuning knob combined with conventional electrostatic gating.

Substrate and crumple structural effects on plasmonic resonances

Next, we investigated substrate-induced effects on the plasmonic resonances in uniaxially crumpled graphene structures. Figure 3a shows the optical absorption spectrum of free-standing crumpled graphene in comparison to that of crumpled graphene on an undulating substrate (for a comparison to crumpled graphene on a flat substrate, see Materials and methods and Fig. S3a). The λ_{res} of the crumpled graphene on the undulating substrate was redshifted compared to that of the free-standing crumpled graphene (5.065 μm vs. 4.457 μm). The redshift of λ_{res} is attributed to confinement of resonant plasmons exclusively in the apex of the crumpled graphene on the undulating substrate (the upper left inset of Fig. 3a). It is notable that the field intensity distribution profiles in the apex and valley regions are different because of the broken symmetry of the plasmonic resonance modes between the apex and valley due to the undulating substrate^{9,36}. The σ_{ext} and quality factor (Q -factor) were enhanced approximately twofold for the undulating substrate compared to the free-standing crumpled graphene (14.0 vs. 7.6% and 63.8 vs. 26.1) (Fig. S3b). However, the crumpled graphene supported on a flat substrate exhibited plasmonic resonances similar to the free-standing graphene ($\lambda_{\text{res}} = 4.488$ μm vs. 4.457 μm , Q -factors = 26.2 vs. 26.1, and $\sigma_{\text{ext,max}} = 6.9$ vs. 7.6%) (Fig. S3a, b). These results show that the refractive index of the substrate enables control of the charge interactions on confronting

graphene surfaces. Therefore, rational choice and design of substrates enables further modulation of plasmonic resonances in mechanically reconfigurable crumpled graphene structures.

To further explore the shape-induced plasmonic resonance of crumpled graphene, we investigated plasmonic resonances in biaxially crumpled graphene (Fig. 3b). We studied a biaxially crumpled graphene structure with the same crumpled wavelengths in both crumpling directions ($\lambda_{c,x} = \lambda_{c,y}$) (the bottom inset of Fig. 3b). The biaxial crumpling of graphene enabled a twofold stronger coupling of the incident light to plasmonic resonance modes (i.e., higher $\sigma_{\text{ext}} \sim 1$ vs. ~ 0.5) (Fig. S4b) and an enhancement of the near-field intensity by a factor of $1.5^2 = (60/40)^2$ compared to uniaxially crumpled graphene (the upper left insets of Fig. 3b vs. the inset of Fig. S4a). Moreover, the biaxially crumpled graphene showed strong near-field enhancements on the slope (the upper left inset of Fig. 3b), whereas the uniaxially crumpled graphene exhibited strong near-field enhancements in the apex and valley regions (the inset of Fig. S4a). With varying h/λ_c from 0.5 to 2 for a constant λ_c , the biaxially crumpled graphene yielded one third the tunable wavelength range of the plasmonic resonance ($\Delta\lambda_{\text{res}} \sim 1.7$ μm) for the uniaxially crumpled graphene ($\Delta\lambda_{\text{res}} \sim 6.1$ μm) (Fig. S4a). Notably, biaxially crumpled graphene exhibited optical absorption reaching up to ~ 0.46 (Fig. 3b) and Q -factors on the order of those in uniaxially crumpled graphene (Fig. S4c). The absorption spectra for biaxially crumpled graphene showed the same plasmonic resonance peak wavelengths at different mobilities, including at 1000, 4000 and 10,000 $\text{cm}^2/(\text{V}\cdot\text{s})$ (Fig. S5a). The Q -factors increased with an increase in carrier mobility (Fig. S5a; the inset of Fig. 3b vs. Fig. S5b). The biaxially crumpled graphene shows a smaller range for wavelength tunability than the uniaxial case but shows enhanced far-field coupling of the incident light for inducing plasmonic resonances, which can be useful for ultrasensitive plasmonic detection.

Plasmonic resonances of finite-area crumpled graphene flakes

Since our studies up to this point have only dealt with infinite sheets of graphene, we explored finite-area biaxially crumpled graphene flakes patterned into hundreds of nanometer-size squares. Such crumpled graphene flakes may find use in bio-imaging and sensing. Figure 4a shows the extinction cross-section spectra for a free-standing biaxially crumpled graphene flake with three periods of crumples in both the x and y directions. Plasmonic resonance peaks were observed for various values of h/λ_c . The near-field distributions at the plasmonic resonance peaks show that resonant plasmons were strongly confined to the center of the crumpled graphene

Fig. 3 Substrate effect for uniaxially crumpled graphene structures and plasmonic resonances for biaxially crumpled graphene. a Optical absorption spectra of the uniaxially crumpled graphene structures with a substrate and without a substrate at $\lambda_c = 100$ nm, $h/\lambda_c = 1$, $E_F = 0.64$ eV, and $\mu = 1000$ cm²/(V·s). The insets show schematic illustrations of the uniaxially crumpled graphene with and without a substrate together with the near-field distributions normalized by E_0 at the plasmonic resonance wavelengths λ_{res} . **b** Maximum optical absorption values $\sigma_{abs,max}$ of uniaxially and biaxially crumpled graphene structures at the plasmonic resonance wavelengths for various h/λ_c from 0.5 to 2 at $\lambda_c = 250$ nm ($E_F = 0.64$ eV and $\mu = 10,000$ cm²/(V·s)). The insets show optical absorption spectra (upper right) for various h/λ_c and a schematic illustration (bottom) of the biaxially crumpled graphene structures. The upper left insets show normalized near-field distributions ($|E|/|E_0|$) for the biaxially crumpled graphene structures (x - z and y - z cross-sectional planes for $h/\lambda_c = 1$)

flake (the insets of Fig. 4a). The biaxially crumpled graphene flake exhibited near-field intensity enhancements of $\sim 1 \times 10^4$, compared to $\sim 1.5 \times 10^3$ for patterned graphene nanostructures, such as nanodisk dimers³⁷. We note that the extinction cross-section spectra with $\mu = 10,000$ cm²/(V·s) showed the same plasmonic resonance peak wavelengths as those with lower mobilities, including at 1000 and 4000 cm²/(V·s) (Fig. S6a). Crumpled graphene flakes may provide several benefits, including enhanced coupling with an external light and strong near-field enhancements.

Finally, to investigate the light-matter interactions^{38,39} of crumpled graphene flakes, we introduce a dipole

emitter placed 40 nm above the center of a free-standing biaxially crumpled graphene flake. The orientation of the dipole moment for the emitter is parallel to the x crumpling direction (the inset of Fig. 4b). To characterize the decay rate enhancement by crumpled graphene flakes, we obtained radiative decay rates (Γ_{rad}) and total decay rates (Γ), which were normalized by those of the emitter in free space without the crumpled graphene flake (Γ_0). Figure 4b shows the radiative decay rate spectra for various values of h/λ_c . We observed significant enhancements in the radiative decay rates, up to three orders of magnitude, at the plasmonic resonance peaks (Fig. 4b) as well as maximum Purcell factors (Γ/Γ_0) of $\sim 1.6 \times 10^5$ (Fig. S7). Our

Fig. 4 Plasmonic resonances for finite-area biaxially crumpled graphene flakes. **a** Optical extinction cross-section of the finite-area biaxially crumpled graphene structures with three periods of crumples with varying h/λ_c from 0.5 to 1.0 at $\lambda_c = \lambda_{c,x} = \lambda_{c,y} = 250$ nm. Normal incident light illuminated the top of the biaxially crumpled graphene flake along the z direction. The polarization of the incident light was along the crumples in the x direction. The insets show normalized near-field distributions in the biaxially crumpled graphene flakes for various h/λ_c . **b** Radiative decay rates of the dipole emitter placed 40 nm above the center of a biaxially crumpled graphene flake for various h/λ_c from 0.5 to 1.0 at $\lambda_c = 250$ nm, $E_F = 0.64$ eV, and $\mu = 10,000$ $\text{cm}^2/(\text{V}\cdot\text{s})$. The inset shows a schematic illustration for a biaxially crumpled graphene flake with a dipole emitter. The dipole moment orientation for the emitter lies parallel with the x crumpling direction. **c** Radiative decay rates as a function of vertical distance (z) between a dipole emitter and a biaxially crumpled graphene flake at the plasmonic resonances. h/λ_c varied from 0.5 to 1.0, with $\lambda_c = 250$ nm, $E_F = 0.64$ eV, and $\mu = 10,000$ $\text{cm}^2/(\text{V}\cdot\text{s})$. The inset shows a schematic illustration for a biaxially crumpled graphene flake with a dipole emitter placed at various vertical distances (z)

maximum Purcell factor was similar to that previously reported for graphene disks⁴⁰. The radiative decay rates at λ_{res} increased by an order of magnitude from 0.6×10^3 to 5.6×10^3 with increasing h/λ_c from 0.5 to 1. These results show that the reconfiguration of crumpled graphene flakes enables tunability for the decay rate enhancements. We note that the radiative decay rate spectra with $\mu = 10,000 \text{ cm}^2/(\text{V}\cdot\text{s})$ showed the same plasmonic resonance peak wavelengths as those with a lower mobility of $1000 \text{ cm}^2/(\text{V}\cdot\text{s})$ (Fig. S6b). We also observed a change in the radiative decay rate (Fig. 4c) and total decay rate (Fig. S8) for various vertical distances from the emitter to the crumpled graphene flake between 0 and 120 nm at the plasmonic resonances. The radiative decay rates showed enhancements of three orders of magnitude over a wide range of vertical distance between 20 and 110 nm. This suggests that crumpled graphene flakes can be used to perform robust molecular sensing against the locational uncertainty of molecules. The plasmonic resonances at mid-infrared wavelengths of $\sim 6 \mu\text{m}$ together with the large Purcell factors as well as the tunability of the plasmonic resonances show potential for ultrasensitive plasmonic detection of molecules⁴¹.

Conclusions

In summary, we have demonstrated, for the first time, mechanically reconfigurable crumpled graphene exhibiting broadband tunability of strong plasmonic resonances from the mid- to near-infrared. The reconfigured crumpled graphene structures, with varying crumple wavelengths and heights, enabled the manipulation of plasmonic resonances over a tunable wavelength range five times broader than that afforded by conventional electrical tuning of graphene. We have also demonstrated efficient excitation of graphene plasmons in crumpled graphene structures by far-field coupling of light as well as strong confinement of the excited plasmons in crumpled graphene. The strong plasmonic resonances enabled optical absorption enhancements of up to ~ 0.46 as well as high near-field enhancements on the order of 1×10^4 for biaxially crumpled graphene flakes. Furthermore, we showed strong light–matter interactions for the crumpled graphene flakes, which enabled large decay rate enhancements to yield a maximum Purcell factor of $\sim 1.6 \times 10^5$. We believe that our crumpled graphene structures, with strong and broadly tunable plasmonic resonances in the near- to mid-infrared, can find broad applications^{1,42,43}, including photonic and optoelectronic devices, ultrasensitive chemical and biological sensing, photovoltaic devices, and spectroscopy.

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Conflict of interest

The authors declare that they have no conflict of interest.

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