Highlights

Advanced materials and techniques for organic electronics, biomedical and sensing applications - 2019

Multipolar terahertz absorption spectroscopy ignited by graphene plasmons

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Terahertz absorption spectroscopy plays a key role in physical, chemical and biological systems as a powerful tool to identify molecular species through their rotational spectrum fingerprint. Owing to the sub-nanometer scale of molecules, radiation-matter coupling is typically dominated by dipolar interaction. We have shown that multipolar rotational spectroscopy of molecules in proximity of localized graphene structures can be accessed through the extraordinary enhancement of their multipolar transitions provided by terahertz plasmons. In particular, specializing our calculations to homonuclear diatomic molecules, we demonstrate that a micron-sized graphene ring with a nano-hole at the core combines a strong near-field enhancement and an inherently pronounced field localization, enabling the enhancement of the dipole-forbidden terahertz absorption cross-section of ionized molecular hydrogen by 8 orders of magnitude. Our results shed light on the strong potential offered by nano-structured graphene as a robust and electrically tunable platform for multipolar terahertz absorption spectroscopy at the nanoscale.

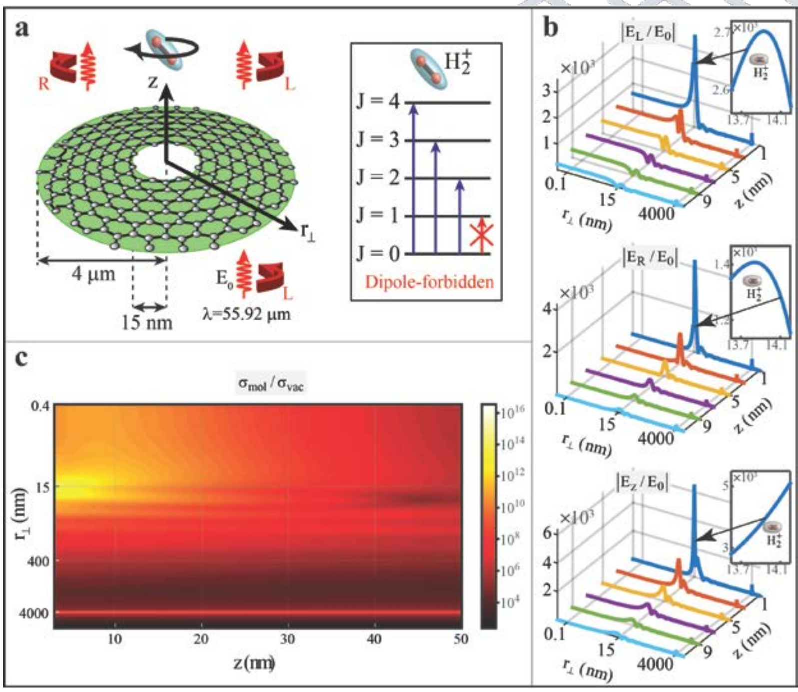

Fig. 1. **a** Graphene micro-ring of radius 4 µm with a nano-hole of radius 15 nm at the core surrounded by dipole-inactive molecules H_2^+. The system is excited by a left-hand (L) circularly polarized THz wave of amplitude E_0 and wavelength λ=55.92 µm. The scattered radiation has both left and right (R) circularly polarized components. **b** Dependence of the scattered fields components E_L, E_R, and E_z over the in-plane radius $r_\perp$ and the altitude z. Note the large field enhancement $|\mathbf{E}|>>E_0$ close to the inner edge at $r_\perp$=15 nm. Note also that the scattered field has deep-subwavelength features whose spatial scale is comparable with the H_2^+ spatial extension (insets). **c** Contour plot of the plasmon-enhanced molecule absorption cross-section σ_{mol} rescaled to its vacuum value σ_{vac} averaged over all molecular orientations as a function of the in-plane radius $r_\perp$ and the altitude z. Note that the normalized absorption cross-section ranges from 10^4 to 10^{16}.