NEMESYS Project- Calibration of the fine-structure constant of graphene by time-dependent density functional theory.

One of the amazing properties of graphene is the ultra-relativistic behavior of its loosely bound electrons, mimicking massless fermions that move with a constant velocity, being inversely proportional to a fine-structure constant \( \alpha_g \) of the order of unity. The effective interaction between these quasi-particles is, however, better controlled by the coupling parameter \( \alpha^* = \frac{\alpha_g}{\epsilon} \), which accounts for the dynamic screening due to the complex permittivity \( \epsilon \) of the many-valence electron system. This concept was introduced in a couple of previous studies [Reed et al., Science 330, 805 (2010), Gan et al., Phys. Rev. B 93, 195150 (2016)], where inelastic x-ray scattering measurements on crystal graphite where converted into an experimentally derived form of \( \alpha^* \) for graphene, over an energy-momentum region on the eV-Å−1 scale. Here, an accurate theoretical framework is provided for \( \alpha^* \), using time-dependent density functional theory in the random phase approximation, with a cut-off in the interaction between excited electrons in graphene, which translates to an effective inter-layer interaction in graphite. The predictions of the approach are in excellent agreement with the above-mentioned measurements, suggesting a calibration method to substantially improve the experimental derivation of \( \alpha^* \), which tends to a static limiting value of \( \sim 0.14 \). Thus, the ab initio calibration procedure outlined demonstrates the accuracy of perturbation expansion treatments for the two-dimensional gas of massless Dirac fermions in graphene, in parallel with quantum electrodynamics.

Summary


Graphene, the two-dimensional (2D) allotrope of carbon with sp2-bonded honeycomb lattice, is the first discovered and currently most studied atomically thin material, due to a variety of potential uses. On the electronic side, the unique properties of graphene stem from its semimetallic band structure around the Fermi energy \( E_F \), with the valence (\( \pi \)) and conduction (\( \pi^* \)) energy levels exhibiting a conical dispersion vs the in-plane momentum at the six corners of the (hexagonal) first Brillouin zone (1st BZ), i.e., the Dirac points. This peculiar feature has allowed the development of a quasi-particle description of charge transport, consisting of charge-carriers that behave as massless Dirac fermions on a velocity scale, characterized by the group velocity of the \( \pi \) and \( \pi^* \) electrons at the Dirac points, i.e., the Fermi velocity \( v_F \sim c/300 \), with \( c \sim 137 \) being the velocity of light in atomic units. Nonetheless, the quasi-particle interaction in this picture depends on a (bare) effective fine-structure constant \( \alpha_g = \frac{1}{\sqrt{v_F} c} \sim 2.2 \), being much larger than the vacuum fine-structure constant \( \alpha = \frac{1}{c} \) of quantum electrodynamics (QED). Indeed, many-body corrections to \( v_F \) can significantly lower \( \alpha_g \), which, however, contrary to QED, remains too large for perturbation treatments. On the other hand, \( \alpha_g \) is too small for strong-coupling approaches. Attempts to reduce \( \alpha_g \) by changing the supporting dielectric medium lead to \( \alpha^* = \frac{\alpha_g}{\epsilon} \), where \( \epsilon \) is the constant permittivity of the ‘background’ that embeds the graphene sheet. Even in this case, \( \alpha^* \) remains not far from unity. A more complete ‘view’ of the interaction strength between the band electrons in graphene amounts to replacing \( 1/\epsilon \) in the Coulomb interaction between excited electrons within the graphene sheet, which reflects in the interlayer interaction in graphite. A substantial improvement on the determination of \( \alpha^* \) is presented, suitable for transfer-red energies from the far-infrared to the extreme ultraviolet, and in-plane momenta up to \( \sim 1 \ \text{Å}^{-1} \). An exploration on the small in-plane momentum region yields the static limiting value \( \alpha^* \sim 0.14 \), in agreement with [1], which supports the idea that the massless Dirac fermions of graphene experience a suf-
ficiently weak interaction. More importantly, a procedure to extrapolate the effective fine-structure constant of Dirac-cone materials is outlined.