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Harnessing ultraconfined graphene plasmons to probe the electrodynamics of superconductors

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We show that the Higgs mode of a superconductor, which is usually challenging to observe by far-field optics, can be made clearly visible using near-field optics by harnessing ultraconfined graphene plasmons. As near-field sources we investigate two examples: graphene plasmons and quantum emitters. In both cases the coupling to the Higgs mode is clearly visible. In the case of the graphene plasmons, the coupling is signaled by a clear anticrossing stemming from the interaction of graphene plasmons with the Higgs mode of the superconductor. In the case of the quantum emitters, the Higgs mode is observable through the Purcell effect. When combining the superconductor, graphene, and the quantum emitters, a number of experimental knobs become available for unveiling and studying the electrodynamics of superconductors.

Significance

Superconductivity and plasmonics constitute two extremely vibrant research topics, although with often nonoverlapping research communities. Here, we bridge these two active research fields by showing that graphene plasmons’ unprecedented light localization into nanometric scales can be exploited to probe the electrodynamics (including collective excitations) of superconductors. Our findings are important both from a fundamental standpoint, representing a paradigm shift (i.e., probing of Higgs modes by light fields), and also for future explorations interfacing nanophotonics with strongly correlated matter, which holds prospects for fostering additional concepts in emerging quantum technologies.


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through the existence of an anticrossing-like feature in the near-field reflection coefficient. Furthermore, the energy and wave vector associated with this feature can be continuously tuned using multiple knobs, e.g., by changing 1) the temperature of the superconductor, 2) the Fermi level of the graphene sheet, or 3) the graphene–superconductor separation.

Finally, we suggest an alternative observation of the GPs–Higgs coupling through the measurement of the Purcell enhancement (23, 33, 34) near the heterostructure. To that end, we calculate the electromagnetic local density of states (LDOS) above the graphene–dielectric–superconductor heterostructure; our results show that, in the absence of graphene, the coupling between the superconductor’s surface polariton and its Higgs mode leads to an enhancement of the LDOS near the frequency of the latter. The presence of graphene changes qualitatively the behavior of the decay rate around the frequency of the Higgs mode, depending strongly on the emitter–graphene distance.

Theoretical Background

Electrodynamics of Bardeen–Cooper–Schrieffer–Like Superconductors. The electrodynamics of superconductors and other strongly correlated matter constitute a fertile research area (29, 30). In the following, we assume that the superconducting material is well described by the Bardeen–Cooper–Schrieffer (BCS) theory of superconductivity (21, 35, 36). Chiefly, the microscopically derived linear optical conductivity tensor of a superconductor requires a nonlocal framework due to the finiteness of the Cooper-pair wave function. For homogeneous superconducting media, the longitudinal and transverse components of the nonlocal optical conductivity tensor—while treating nonlocality to leading order—can be expressed as (21, 37, 38)

\[
\sigma_L(q, \omega) = \sigma_D(\omega) \frac{1}{1 - 3\tilde{\alpha}(\omega, T)(\frac{q}{a})^2}, \quad \sigma_T(q, \omega) = \sigma_D(\omega) \left[ 1 + \tilde{\alpha}(\omega, T)(\frac{q}{a})^2 \right],
\]

respectively, where \( \sigma_D(\omega) = \frac{\text{im} \sigma_L(q, \omega)}{\text{im} \sigma_T(q, \omega)} \) is the Drude-like conductivity, and the dimensionless coefficient \( \tilde{\alpha}(\omega, T) \) amounts to

\[
\tilde{\alpha}(\omega, T) = \frac{\hbar^4}{30\pi^2 n m^3 c^2} \int_0^\infty \! dk \, k^6 \times \left\{ 2f(E_k)[1 - f(E_k)] \left[ 1 - \frac{\Delta_k^2(T)}{E_k^2} \right] + \frac{(\hbar\omega)^2 \Delta_k^2(T)}{\hbar\omega^2} \frac{1 - 2f(E_k)}{(2E_k)^2} \right\}.
\]

In the previous expression, \( E_k = \sqrt{(\epsilon_k - \mu)^2 + \Delta_k^2(T)} \) is the quasiparticle excitation energy at temperature \( T \), where \( \mu \simeq E_F = \frac{\hbar^2}{2m}(3\pi^2 n)^{2/3} \) is the superconductor’s chemical potential, \( \epsilon_k = \hbar^2 k^2/2m \) is the single-particle energy of an electron with wave vector \( k \), \( \Delta_k(T) \equiv \Delta_k(0)(T) = 1.76 \times k_B T_c(1 - (T/T_c)^4)^{1/2} \Theta(T_c - T) \) is the temperature-dependent gap parameter of the superconductor, and \( f(E_k) = [\exp(E_k/k_B T) + 1]^{-1} \) is the Fermi–Dirac distribution.

In possession of the response functions epitomized by Eq. 1, we employ the semiclassical infinite barrier (SCIB) formalism (23, 39) to describe electromagnetic phenomena at a planar dielectric–superconductor interface (37, 38, 40). Within this framework, the corresponding reflection coefficient for \( p \)-polarized waves is given by (SI Appendix) (23, 39)

\[
r_{sc}^p = \frac{k_{z,d} - \epsilon_d \Xi}{k_{z,d} + \epsilon_d \Xi},
\]

with \( k_{z,d} = \sqrt{(\epsilon_d)^2 - q_{||}^2} \), and \( \Xi \) has the form

\[
\Xi = \frac{i}{\pi} \int_{-\infty}^{\infty} \! dq_{\perp} \left[ \frac{q_{\perp}^2}{\epsilon_1(q, \omega)} + \frac{q_{\perp}^2}{\epsilon_T(q, \omega) - (\Xi)^2} \right],
\]

where \( q_{||} = \sqrt{q_{\perp}^2 + q_{\perp}^2} \), and \( \epsilon_{LT} = \epsilon_{\infty} + i\epsilon_{LT}/(\omega\epsilon_0) \) are the components of the superconductor’s nonlocal dielectric tensor (we take \( \epsilon_{\infty} = 1 \) hereafter).

In what follows, we assume a typical high-\( T_c \) superconductor, such as yttrium barium copper oxide (YBCO), with a normal state electron density of \( n = 6 \text{ nm}^{-3} \) and a transition temperature of \( T_c = 93 \text{ K} \) (yielding a superconducting gap of \( \Delta_0(0) \approx 14.2 \text{ meV} \) (37, 38, 41).

Electrodynamics in Graphene–Dielectric–Superconductor Heterostructures. With knowledge of the reflection coefficient for the dielectric–superconductor interface (2), the overall reflection coefficient, i.e., that associated with the dielectric–graphene–dielectric–superconductor heterostructure, follows from imposing Maxwell’s boundary conditions (42) at all of the interfaces that make up the layered system. At the interface defined by the two-dimensional graphene sheet, the presence of graphene enters via a surface current with a corresponding surface conductivity (22).

Signatures of the system’s collective excitations can then be found by analyzing the poles of the corresponding reflection coefficient, which are identifiable as features in the imaginary part of the (overall) reflection coefficient, \( \text{Im} r_p \) (SI Appendix).

Coupling of the Higgs Mode of a Superconductor with Graphene Plasmons

Signatures of the Higgs Mode Probed by Graphene Plasmons. Like ordinary conductors (44), superconductors can also sustain surface plasmon polaritons (SPPs) (45, 46). In turn, these collective excitations can couple to the superconductor’s Higgs mode.
Typically such interaction is extremely weak due to the large mismatch between the superconductor’s plasma frequency, $\omega_p$, and that of its Higgs mode, $\omega_H = 2\Delta_0/\hbar$; for instance, $\omega_H/\omega_p \sim 10^{-2}$, with $\omega_p$ and $\omega_H$ falling, respectively, in the visible and terahertz spectral ranges. As a result, at frequencies around $\omega_H$ the SPP resembles light in free space and thus the SPP–Higgs coupling is essentially as weak as when using far-field optics (Fig. 2A).

On the other hand, graphene plasmons not only span the terahertz regime but also attain sizable plasmon wave vectors at those frequencies (22, 23). Moreover, when the graphene sheet is near a metal—or a superconductor for that matter—graphene’s plasmons become screened and acquire a nearly linear (acoustic) dispersion, pushing their spectrum further toward lower frequencies (i.e., a few terahertz) and larger wave vectors (23–27). Therefore, these properties of acoustic-like GPs can be harnessed by placing a graphene monolayer near a superconducting surface, thereby allowing the interaction of graphene’s plasmons with the Higgs mode of the underlying superconductor (Fig. 2B). In this case the plasmon–Higgs interaction is substantially enhanced, a fact that is reflected in the observation of a clear anticrossing in the GP’s dispersion near $\omega_H$, which, crucially, is orders of magnitude larger than that observed in the absence of graphene (Fig. 2A and B).

Furthermore, the use of graphene plasmons for probing the superconductor’s Higgs mode comes with the added benefit of control over the plasmon–Higgs coupling by tuning graphene’s Fermi energy electrostatically (22, 23, 47–49). This is explicitly shown in Fig. 3A, for a vacuum–hexagonal boron nitride (hBN)–graphene–hBN superconductor heterostructure; as before, the manifestation of the Higgs mode near the superconductor’s Higgs mode; here, $\Delta q = 0$. Typically such interaction is extremely weak due to the large mismatch between the superconductor’s plasma frequency, $\omega_p$, and that of its Higgs mode; $\omega_H = 2\Delta_0/\hbar$; for instance, $\omega_H/\omega_p \sim 10^{-2}$, with $\omega_p$ and $\omega_H$ falling, respectively, in the visible and terahertz spectral ranges. As a result, at frequencies around $\omega_H$ the SPP resembles light in free space and thus the SPP–Higgs coupling is essentially as weak as when using far-field optics (Fig. 2A).

Higgs Mode Visibility through the Purcell Effect. One way to overcome the momentum mismatch and investigate the presence of electromagnetic surface modes is to place a quantum emitter (22, 51–53) (herein modeled as a point-like electric dipole) in the proximity of an interface and study its decay rate as a function of the emitter–surface distance. With the advent of atomically thin materials, and hBN in particular, all of the relevant distances, i.e., emitter–superconductor, emitter–graphene, and graphene–superconductor, can be tailored with nanometric precision [e.g., by controlling the number of stacked hBN layers (each $\sim 0.7$ nm thick) (25, 32) or using atomic layer deposition (54, 55)]. Although the availability of good emitters in the terahertz range is unarguably limited, semiconductor quantum dots with intersublevel transitions in this range and with relatively long relaxation times do exist (56). The modification of the spontaneous decay rate of an emitter is a repercussion of a change in the electromagnetic LDOS, $\rho(R)$, and it is known as the Purcell effect (23, 33, 34). Specifically, the Purcell factor—defined as the ratio $\rho(R)/\rho(0)$, where $\rho(0)$ is the LDOS experienced by an emitter in free space—can be greatly enhanced by positioning the emitter near material interfaces supporting electromagnetic modes (which are responsible for augmenting the LDOS). In passing, we note that this LDOS enhancement does not strictly require an “emitter,” since it can also be probed through the interaction of the sample with the illuminated tip of a near-field optical microscope (which may be modeled as an electric dipole in a first
Here, we have modeled hBN’s optical properties using a dielectric tensor of the form Drude damping. The thickness of the bottom hBN slab is given by $t_B$.

Then, the orientation-averaged Purcell factor—or, equivalently, the LDOS enhancement—can be determined via (34)

$$\frac{\rho(z)}{\rho_0} = 1 + \frac{1}{2} \int_0^\infty ds \text{Re} \left( \left( \frac{s_3}{s_0} - s_0 s_3 \right) r_p e^{2i\pi s_0 z} \right),$$  \[3\]

where $s_i = \sqrt{1 - s^2}$, with $s = q_0 e / \omega$ denoting a dimensionless in-plane wave vector, and $z = d - t'$ is the vertical coordinate relative to the surface of the topmost hBN layer, and where $d$ is the emitter–graphene distance.

Fig. 4 shows the LDOS enhancement experienced by an emitter (or a nanosized tip) in the proximity of a superconductor; Fig. 4A, B, D, and E refers to the case in the presence of graphene (located between the superconductor and the emitter), whereas Fig. 4C depicts a scenario where the graphene sheet is absent. The graphene sheet modifies the LDOS, affecting not only the absolute Purcell factor but also the peak/dip feature around the energy of the Higgs mode, $\hbar\omega_{H} = 2\Delta_c$. Such modification depends strongly on the emitter–graphene separation $d$ (Fig. 4A and B). Fig. 4D shows the LDOS enhancement for $T > T_c$ (i.e., above the superconductor’s transition temperature) and thus the feature associated with the Higgs mode vanishes; all that remains is a relatively broad feature related to the excitation of graphene plasmons.

Finally, Fig. 5 depicts the LDOS enhancement for different values of graphene’s Fermi energy (which can be tuned electrostatically), for two fixed emitter–graphene distances: $d = 13$ nm (Fig. 5, Top row) and $d = 2$ nm (Fig. 5, Middle row). For weakly doped graphene and the larger $d$ the sharp feature associated with the hybrid GPs–Higgs mode dominates the Purcell factor, being eventually overtaken by the broader background with increasing $E_p^G$. To unveil the mechanisms underpinning the LDOS enhancement, we plot in Fig. 5, Bottom row the $q_z$-space differential LDOS enhancement (tantamount to the so-called $q_z$-space power spectrum, 39), which amounts to the integrand of Eq. 3. In the near field (well realized for the chosen setup and parameters), there are two contributions (34, 39): one from a resonant channel, corresponding to the excitation of the coupled Higgs–GP mode, and a broad, nonresonant contribution at larger $q_z$ due to lossy channels (phenomenologically incorporated through the relaxation rates $\gamma, \gamma_p$).

Mathematically, the polariton (Higgs–GP mode) resonant contribution arises from the pole in $\text{Im} r_p$, occurring at $q_0 \simeq \text{Re} q_{GP}(\omega)$ (where $q_{GP}(\omega)$ is the wave vector of the Higgs–GP mode at frequency $\omega$ that satisfies the dispersion relation) (Fig. 3). Consistent with this, the peak associated with the Higgs–GP polariton contribution to the $q_z$-space differential LDOS occurs at a larger wave vector in the $E_p^G = 50$ meV case, since, for the same frequency, the Higgs–GP dispersion shifts toward larger wave vectors upon decreasing $E_p^G$ (24, 28). Ultimately, the amplitude of the resonant contribution depends on the specifics of the dispersion relation (i.e., $q_{GP}(\omega) = \text{Re} q_{GP}(\omega) + i \text{Im} q_{GP}(\omega)$) and is further weighted by the $\exp (-2q_0 z)$ factor that depends not only on the peak’s location, $q_0(\omega) \simeq \text{Re} q_{GP}(\omega)$ (and whose width $\propto \text{Im} q_{GP}(\omega)$), but

**Fig. 3.** Tuning the hybridization of acoustic-like plasmons in graphene with the Higgs mode of a superconductor in air–hBN–graphene–hBN–superconductor heterostructures. The colormap indicates the loss function via $\text{Im} r_p$. (A and B) Spectral dependence upon varying the Fermi energy of graphene (A) and the graphene–superconductor distance (B). Setup parameters: The parameters of the superconductor are the same as in Fig. 2, and the same goes for graphene’s Drude damping. The thickness of the bottom hBN slab is given by $t$, whereas the thickness of the top hBN slab, $t'$, has been kept constant ($t' = 10$ nm). Here, we have modeled hBN’s optical properties using a dielectric tensor of the form $\varepsilon_{\text{hBN}} = \text{diag}(\varepsilon_{xx}, \varepsilon_{yy}, \varepsilon_{zz})$ with $\varepsilon_{xx} = \varepsilon_{yy} = 6.7$ and $\varepsilon_{zz} = 3.6$ (24, 49, 50).
also on the emitter’s position \( z = d - t' \) (Eq. 3). Finally, we stress that the relative contribution of each of the above-noted decay channels is strongly dependent on the emitter–graphene distance \( d \) (with the nonresonant, lossy contribution eventually dominating at sufficiently small emitter–graphene separations—quenching) (34, 39).

**Conclusion and Outlook**

We have shown that signatures of a superconductor’s Higgs mode can be detected by exploiting ultraconfined graphene plasmons supported by a graphene sheet placed in a superconductor’s proximity. In particular, the presence of the Higgs mode for \( T < T_c \) can be readily identified through an anticrossing feature that attests to the coupling between graphene plasmons and the superconductor’s Higgs mode. Further, we suggest that the excitation of the Higgs mode of superconductors could also be detected through the emergence of a peak or a dip in the near-field’s Purcell factor and whose shape (peak or dip) depends on the coupling between the emitter and the continuum of the hybrid GP–Higgs mode. This coupling is most efficient for small Fermi energies and short distances between the superconductor and the emitter.

Experimentally, the GP–Higgs interaction can be investigated using state-of-the-art cryogenic scanning near-field optical microscopy (SNOM) (43). Alternatively, more conventional spectroscopies relying on far-field optical techniques can also be explored by nanopatterning the graphene itself (e.g., into ribbon arrays) or its nearby materials (for example, the hBN or the superconductor). Examples of the latter—which have the benefit of preserving graphene from nanofabrication-induced defects—include the configurations studied in refs. 25 and 32, while the former approach can still be pursued using cutting-edge electron-beam lithography (61). Another possibility is the use of highly localized, local back-gate-free graphene doping modulation by placing a pristine graphene sheet on a substrate with patterned \( \alpha \)-RuCl\(_3\) (62).

Finally, there are a number of open questions that can spur from this work; e.g., if conductive thin films were added in direct electrical contact with the superconductor, then bound Andreev quasiparticle states inside the superconducting energy gap can form, being solutions to the Bogoliubov–de Gennes equations (63). Another enticing outlook is the prospect of using highly confined GPs for investigating Josephson plasma waves in layered high-\( T_c \) superconductors (29, 64, 65). The present formalism could be extended to the coupling of the above-noted types of modes (although this likely requires the use of more sophisticated models beyond the SCIB model employed here).

The work presented here sheds light on the fundamentals of collective excitations in architectures containing two-dimensional materials and superconductors and constitutes a proof-of-principle proposal, paving the way for prospective experimental investigations into the electrodynamics of superconductors using ultraconfined graphene plasmons.

**Data Availability.** All study data are included in this article and/or SI Appendix.

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An Introduction to Graphene Plasmonics


