15. Nonlocality in polar dielectrics

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Current status

Phonon polaritons, hybrid modes formed by the strong coupling of electromagnetic fields with optical phonons in polar crystals, have emerged as a powerful platform for sub-diffractional mid-infrared (mid-IR) photonics. Unlike plasmons, which rely on free carrier oscillations and can suffer from high losses, phonon polaritons benefit from the intrinsic low-loss nature of lattice vibrations. As such, they have found application in a range of areas including sensing, nonlinear optics and thermal emitters [347]. The standard approach to modeling polar crystals relies on a local dielectric approximation, which implicitly assumes a negligible momentum dependence of the phonon response. This local treatment works well at larger feature sizes, as in bulk transverse optical (TO) and longitudinal optical (LO) phonons have well-defined frequencies at optical wavelengths, separated by the Reststrahlen band. However, at the nanoscale, extreme light confinement can probe the phonon dispersion well inside the Brillouin zone, phonons acquire a non-negligible wavevector dependence, and their propagation within a confined geometry cannot be neglected. Under these conditions nonlocal effects, where the material response at a given point depends on fields in a finite surrounding region, become significant. Studying the electrodynamics of the system requires then taking into account the phononic, mechanical degrees of freedom on-par with the electromagnetic ones. This boils down to the Poynting vector, describing energy fluxes, acquiring a second term, proportional to the stress tensor of the lattice and describing energy propagating in the form of elastic energy in the solid [348]. Imposing the continuity of such a generalized Poynting vector allows one to determine boundary conditions on the mechanical fields that, together with the usual Maxwell boundary conditions, make it possible to fully solve the coupled light-matter problem. One important feature of these boundary conditions is that they mix longitudinal and transverse degrees of freedom, leading to a modified optical response and the emergence of novel excitations known as longitudinal-transverse polaritons (LTPs), which mix both transverse and longitudinal character. While nonlocal effects are known to be present also in plasmonic systems, and some consequences of nonlocal phenomenology are similar in plasmonic and phononic systems, the origin and consequences of nonlocality in those two leading nanophotonic platforms is fundamentally different, as schematically shown in Fig. 19 [306]. Plasmons disperse toward the blue and longitudinal plasmons, which only exist above the plasma frequency, cannot thus hybridize with electromagnetically localized plasmon-polariton modes, which instead exist only below the plasma frequency where the dielectric function of the metal is negative. Their interaction is necessarily an evanescent phenomenon, leading to nonlocal phenomenology in plasmons being limited to an Ångström-thin skin depth. Optical phonons disperse instead toward the red, making the coupling between longitudinal and transverse degrees of freedom a resonant, propagative phenomenon, leading to the emergence of LTPs, and enhancing by of orders of magnitude the length scales on which nonlocal phenomena can be observed and exploited. The existence of LTPs have been theoretically predicted and experimentally observed in silicon carbide (SiC) nanopillar arrays [349] and in crystal hybrids, polar superlattices with nanometric-sized layers in which standard dielectric modeling fails to reproduce even the qualitative features of experimental reflectance [306]. This success highlights that nonlocality in phonon polariton systems is not a theoretical curiosity but a practical concern that can affect device design and operation. Other predicted impacts of nonlocality on phonon polariton systems are an increased linewidth of Fröhlich resonances in dielectric nanoparticles [306] and the reduction of field confinement in nanogap resonators [350], as in both cases energy can leak out via emission of LO phonons.

Challenges and opportunities

As the length scales of photonic devices approach the phonon propagation length, phonon nonlocality introduces several fundamental and practical challenges. The first and perhaps most direct challenge lies in accurate theoretical modeling. Traditional modeling approaches rely on local dielectric functions fitted to bulk optical constants, which fail to capture the momentum-dependent response of optical phonons. As a result, standard simulation tools and design methodologies yield inaccurate predictions of device resonance frequencies, confinement factors, and field enhancements. This discrepancy complicates the design of ultra-thin and deeply subwavelength structures where the effects of nonlocality are most pronounced. Multiple approaches have been explored, taking explicitly the phononic field into account either in finite-element method (FEM) simulations [350] or in scattering-matrix codes [351], relying on effective medium theories [352], or developing analytical mappings to well-known quantum optics models [353]. Still, these methods have been applied to relatively simple materials and

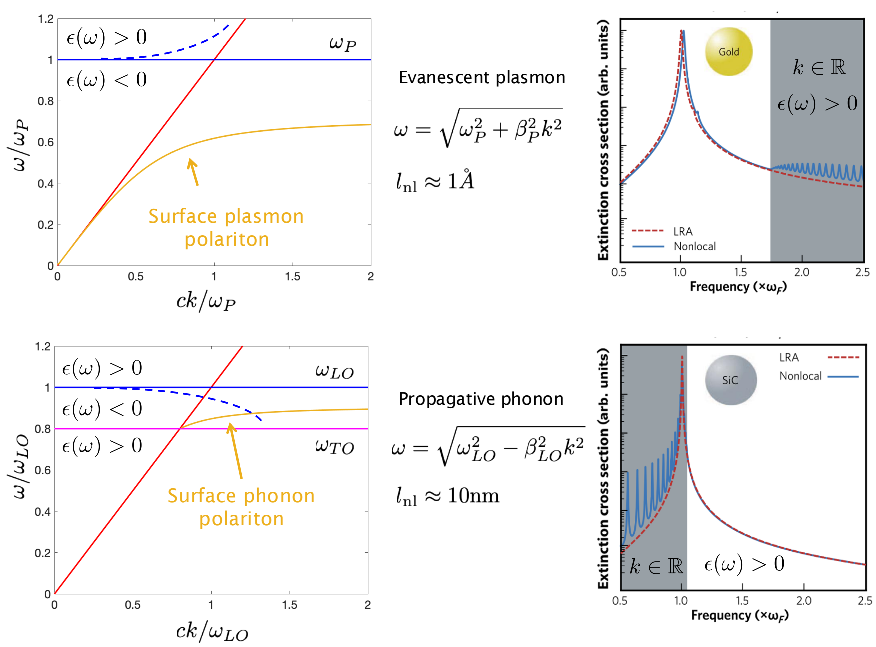

Fig. 19. Comparison of nonlocality in plasmon-polariton (top) and phonon-polariton (bottom) systems. On the left column we show a sketch of the surface polariton dispersions (yellow solid line) with highlighted the regions in which the Drude (top) and Lorentz (bottom) dielectric functions change sign. The red solid lines are the light line and the horizontal lines mark the plasma frequency (P) and the LO and TO phonon frequencies. The dashed blue lines sketch the dispersions of the longitudinal modes, written in the quadratic approximation in the central column for each case. On the right column we show the Fröhlich resonance, of frequency ω_F, for a gold (top) and SiC (bottom) nanosphere, with the extinction cross section calculated using a local (red dashed line) and nonlocal (blue solid line) theory. The shaded region corresponds to the frequency region spanned by the dispersive mode for real values of the wavevector k. Only for phonon-polaritons, due to the red-dispersion of the LO phonon, such a region overlaps with the region of negative dielectric function in which the Fröhlich resonance can exist, leading to resonant energy exchange and to a substantially larger nonlocal length scale $\ell_{\rm nl}$. Data adapted from Ref. [306].

interfaces, with the most complex case a single asymmetric Reststrahlen band in Ref. [351]. For more complex and potentially highly anisotropic materials, in which multiple phonon modes are present in overlapping Reststrahlen bands, the resulting set of coupled boundary conditions could be substantially more challenging to solve. Even for simple materials current approaches rely on approximations which limit the theory's overall domain of applicability. For a start, all the theoretical development cited rely on a quadratic approximation of the phonon dispersion. This means that the frequency of each optical phonon mode is written as a function of the wavevector as $\omega(k) = \sqrt{\omega^2(0) - \beta^2 k^2}$, where $\omega(0)$ and β are the zone-center frequency and velocity parameter of the mode. This can be problematic when considering interfaces between materials with Reststrahlen bands with broad overlaps, in which the frequencies provided by the quadratic approximation can diverge significantly from their physical values. While a completely numerical approach that takes as input a parametrization of the phonon dispersion across all the Brillouin zone could in-principle be used to solve the problem in a more general way, this remains an open problem. Moreover, for extremely small nanolayers (1–2 nm), the microscopic crystal structure starts to play a role, and the thickness of the each layer, used a parameter in the dielectric continuum modeling, is not precisely defined. This adds a material-dependent adjustable parameter to the theory which could be fixed (once each material interface) by *ab initio* simulations [306]. A second challenge is related to experimental characterization. Probing nonlocal phonon effects requires techniques capable of exploring few-nanometer scales, where these phenomena manifest. While various mature approaches to near-field microscopy exist, the dependency of the nonlocal effects on the exact dimensions of the objects requires samples with extremely high uniformity, or the capability to probe a single nano-object, which remains to-date challenging. Despite these hurdles, the opportunities that nonlocal phonon effects unlock are significant. Nonlocality offers a pathway to create novel types of hybrid modes like LTPs which combine the radiative nature of transverse fields and the strong interactions with electrical currents of longitudinal phonons, potentially bridging the gap between electrical excitations and far-field mid-IR emission. Such a capability could enable direct electrical pumping of mid-IR phonon polariton modes, offering a route to electrically driven emitters without the need for engineered electronic transitions such as those in quantum cascade structures. In such a scheme, the Fröhlich interaction, responsible for LO-phonon emission in polar dielectrics, can become dressed by the strong interaction crating the LTPs. The scattering of a conduction electron would then resonantly create an LTP which could subsequently decay emitting mid-infrared light in the far-field. While initial theoretical analysis shows this mechanism to be potentially intense enough for practical applications [354], the intrinsically multi-scale nature of the problem makes it difficult to obtain precise results and an experimental verification of the emission mechanism is still missing. Only recently some more advanced non-equilibrium results obtained with a non-equilibrium Green function approach have appeared [355] and still only for simple structures and low values of the applied bias. Additionally, nonlocal responses could be used to enhance control over optical anisotropy and hyperbolicity in polar materials. By carefully designing nanostructured heterostructures, superlattices, and patterned nanophotonic elements, it may be possible to engineer the dispersion of phonons and thus the properties of the resulting polaritons. This could lead to enhanced sensing capabilities, ultra-confined modes with engineered dispersion, and new opportunities for nonlinear optical processes.

Future developments to address challenges

To effectively harness phonon nonlocality, the next years will likely see a concerted effort along two main axes: theoretical modeling and experimental characterization. From a theoretical standpoint, developing comprehensive and user-friendly computational frameworks that include phonon dispersion and satisfy both electromagnetic and mechanical boundary conditions is a priority. Existing macroscopic models, some already demonstrated in simplified geometries, must be extended and made accessible to a broad community. Incorporating nonlocality into commercial electromagnetic simulation packages will be an important milestone, facilitating widespread exploration. Such models should be flexible enough to handle complex anisotropic materials, arbitrary nanostructures, and large-scale integrated photonic devices. On a fundamental level, more intuitive analytical approaches to interpret nonlocal behavior and guide rational design strategies will also prove valuable. Advanced multi-scale modeling of LTP-based electroluminescent devices will have to advance, allowing to better guide the design of prototype devices to gain a first experimental proof of this novel electroluminescence emission channel. Experimentally, there is a need for the characterization of more LTP materials, which will play the double role of both verifying and pushing forward theoretical developments. Spectroscopic characterization of monodisperse polar nanosphere of decreasing dimension could provide evidence for some of the yet unobserved predictions of the nonlocal theory as well as clarifying the behavior of materials below critical nanometer dimensions. Experiments are necessary to try and observe LTP electroluminescence. Only after a first unequivocal observation of this emission channel an effort to optimize the emission and collection efficiency could take place, which in turn will be necessary to ascertain whether LTP electroluminescence is at most an interesting curiosity or if it has the potential to empower a novel generation of mid-infrared optoelectronic devices. Finally, synergy with other fields—such as ultrafast optics, quantum photonics, and topological photonics—may lead to novel concepts and applications. For instance, the interplay of nonlocal phonon effects with coherent control schemes or coupling to quantum emitters (like defects in diamonds, molecular vibrations or quantum wells) could yield unprecedented control over mid-IR light–matter interactions. Similarly, integrating nonlocal phononic elements into topological platforms might provide robust, loss-resistant channels for infrared light. In short, the future developments needed to address the challenges posed by phonon nonlocality involve building a comprehensive toolkit encompassing theory, experiment, and device engineering. These advances will enable the community to fully realize the potential of nonlocal phonon polaritons in creating next-generation mid-IR photonic technologies.

Concluding remarks

Phonon nonlocality represents both a theoretical and technological frontier in mid-infrared photonics. By moving beyond the local dielectric approximation, we acknowledge the true complexity of polar materials and unlock new phenomena such as longitudinal-transverse polaritons. Although this introduces modeling challenges and demands more refined experimental techniques, it could open up the possibility of creating more compact, efficient, and versatile mid-IR devices. The ability to couple electrical currents directly to radiative modes, leverage nonlocal effects for enhanced field confinement, and engineer the phonon dispersion through nanostructuring and heterostructures suggests a rich landscape of future innovations. As theoretical tools mature and integrate with experimental platforms, and as fabrication techniques continue to improve, phonon-based nonlocal photonics will likely play an increasingly important role. Ultimately, this may enable a new generation of mid-IR emitters, detectors, sensors, and nonlinear optical devices that harness the full power of phonon physics and broaden the scope of photonic materials and metamaterials research.

16. Nonlocal effects in graphene plasmonics

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Current status

Plasmons in graphene and graphene-based nanostructures possess extraordinary properties, featuring deeply subwavelength field confinement ($\lambda_p \ll \lambda_0$, where λ_p and λ_0 denote the graphene-