Dissipative and dispersive optomechanics through complex modal volumes

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Despite the several novel features arising from the dissipative optomechanical coupling, such effect remains vastly unexplored due to the lack of a formalism that captures non-Hermiticity in optomechanical systems. In this Letter, we show that a complex modal volume-based perturbation theory is capable of correctly predicting both dispersive and dissipative optomechanical couplings. We validate our model through simulations and also by comparison with experimental results reported in the literature. Finally, we apply this formalism to plasmonic systems, used for molecular optomechanics, where strong dissipative coupling signatures in the amplification of vibrational modes are observed.

Cavity optomechanics has been a very prolific field of research in the past decades [1, 2], with applications in quantum information processing [3], microwave-to-optical signal conversion [4, 5], sensing and precision measurement [6, 7], and as platform for fundamental physics tests [8–10]. Such developments were built upon key properties of microfabricated devices, such as large optical quality factors, small modal volumes, and large spatial overlap between optical and mechanical modes. More recently, nanoplasmonic resonators have also been pointed out as suitable candidates for quantum molecular optomechanics, where unparalleled zero-point dispersive optomechanical couplings (\(g_0/(2\pi) \approx 100\text{ GHz}\)) [11] have been observed and could lead to the observation of optical mechanical strong-coupling in the few-photons regime, even at low Q-factors (\(Q < 30\) [12]).

While most reports to date rely on dispersively coupled optical and acoustic modes, with frequency pulling \(G_\omega = -\gamma_\omega/\Delta\) [13–15], a complete description of the optomechanical interaction must take into account a dissipative coupling \(G_\kappa = \frac{\kappa}{\Delta}\), where a mechanical displacement \(x\) modulates the optical mode linewidth, which can be of intrinsic nature (\(\kappa_i\)) due to absorption and scattering, and/or extrinsic (\(\kappa_e\)), due to coupling to a coherent excitation channel e.g. waveguide (Figs. 1a and b)). Although, various novel features have been predicted using dissipative coupling, such as cooling in the bad-cavity limit regime (\(\kappa/2 \gg \Omega_m\)) [16] and quantum-limited position measurement [17], experimental demonstrations are still scarce [18, 19] and with a lack of a theoretical framework to engine devices with strong dissipative coupling.

In this Letter, we introduce a non-Hermitian perturbation theory based on complex modal volumes [20, 21] to simultaneously evaluate \(G_\kappa\) and \(G_\omega\) in any geometry undergoing small deformations, as is the case of optomechanical resonators. Although we focus on the intrinsic linewidth perturbation case, \(G_\kappa = \frac{\kappa}{\Delta}\), our approach could be generalized to account for extrinsic losses. While the physicality and extent of the complex modal volume concept remain a matter of debate [6, 22], here we help elucidating it by demonstrating its accuracy in three conceptual devices chosen to illustrate distinct aspects of this formulation: a ring resonator interacting with a lossy element; a split-beam nanocavity torque sensor that has been experimentally reported in Ref. [18]; and a nanoparticle-on-a-mirror (NPoM) [23, 24] scheme, where we show that plasmonic nanocavities used in the context of surface or tip-enhanced Raman scattering (SERS or TERS) naturally display large dissipative coupling signatures, which should be carefully evaluated when considering such platform for quantum optomechanics experiments.

The interplay between optical and acoustic modes is typically modeled using perturbation theory. The mechanical mode deformation couples with the optical field through a change in the electric permittivity (\(\Delta\varepsilon\)). In mesoscopic mechanical resonators (dimensions on the order of the optical wavelength), this coupling arises from two main mechanisms: moving boundaries (MB) [25] and photoelasticity (PE) [26]. In the microscopic regime, e.g. molecular optomechanics, molecules are treated as point dipoles interacting with plasmonic modes.

Previous formulations treat only Hermitian (or lossless) optomechanical systems. In this case, optical eigen-
modes \( \tilde{E}_m \) can be derived to be power-orthogonal \[27\], i.e.
\[
\int \tilde{E}_m^* \cdot \epsilon \tilde{E}_n \, dV = C_{n,m} \delta_{n,m},
\]
where \( \epsilon \) is the permittivity tensor of the medium, \( \delta_{n,m} \) the Kronecker delta and \( C_{n,m} \) a normalizing constant with dimensions of energy. The derivation of such expression relies on the Hermitian property that the eigenvalues (i.e., optical frequencies) of the system are real numbers and lead to the usual definition of an optical mode volume \[28\]. In dissipative systems, however, non-Hermitian operators with imaginary eigenvalues arise, disrupting the usual orthogonality relation and optical mode volume. To overcome this problem, we introduce the use of the bi-orthogonal product \[29\] to derive the optomechanical coupling calculations. Such pseudo-inner product has recently attracted interest due to its applications in non-Hermitian light-matter interactions, such as in the calculations of the Purcell factor \[30, 31\], and also in the context of exceptional points \[32–34\].

Bi-orthogonality relations are derived between left \( \bar{E}_m^L \) and right \( \bar{E}_m^R \) eigenvectors of a generalized eigenvalue equation \[35, 36\]. In this framework, orthogonality is recovered through:
\[
\int \bar{E}_m^L \cdot \epsilon \bar{E}_m^R \, dV = C_{n,m} \delta_{n,m},
\]
where a reciprocal medium is assumed, i.e. \( \epsilon = \epsilon^T \) with “\( T \)” denoting transposition. Since the left and right eigenmodes are not complex conjugated pairs, this integral generally results in complex numbers and may, therefore, be associated with a complex modal volume. The latter is typically defined as
\[
V_m = \int \bar{E}_m^L \cdot \epsilon \bar{E}_m^R \, dV / \left( \bar{E}_m^L(\bar{r}_0) \cdot \epsilon(\bar{r}_0) \bar{E}_m^R(\bar{r}_0) \right),
\]
where \( \bar{r}_0 \) is the position chosen for field normalization. Simple relations between \( \bar{E}_n^L \) and right \( \bar{E}_n^R \) can be derived and are shown in section S1 of the Supplemental Material. The relations above remain valid in lossy systems, where eigenmodes are known as quasi-normal modes or QNMs. Issues with the completeness of QNMs are here circumvented by mapping realistic problems in open space into a computational domain bounded by outgoing boundary conditions, emulated by perfectly matched layers (PML) \[37\].

Numerical modeling.— To validate our approach, we consider a toy model composed of two concentric 250 nm thick GaAs rings, as shown in Fig. 2 a). The (inner) ring resonator, which supports traveling-wave type optical modes, is undoped \( \epsilon/\epsilon_0 = n^2 \), where \( n = 3.46 \) and \( \epsilon_0 \) the vacuum permittivity, while its width and radius are picked to be \( \omega_{\text{ring}} = 500 \text{ nm} \) and \( R = 3.5 \mu \text{m} \), respectively. The external ring is heavily doped and its dielectric constant is given by \( \epsilon/\epsilon_0 = n^2 - j \sigma/(\epsilon_0 \omega) \), with conductivity \( \sigma = 5 \times 10^4 \text{ S.m} \), similar to values achieved with ion implantation in semiconductor substrates; in this case, its width is chosen to be much larger than its skin depth, avoiding reflections at the perfect electric conductor boundary used to limit the simulation domain. This example has the advantage of admitting complex eigenvalues regardless of radiation losses, constituting a relevant case for absorption-induced dissipative coupling \[19\].

The inner ring’s mechanical breathing mode \( \Omega_m/(2\pi) \approx 4.1 \text{ GHz} \) is considered, while the external doped structure is kept fixed. For a given gap between the two rings, the optomechanical coupling was evaluated using both complex and real modal volume approaches. Their distance was varied to encompass both high and low optical-Q regimes, as shown in Fig. 2 b), allowing us to probe the domains of validity of both perturbative calculations. The calculated optomechanical coupling rates were then cross-checked against exact simulations that incorporate both MB and PE effects. For that purpose, the optomechanical ring’s geometry is deformed following the mechanical mode profile using COMSOL®’s Moving Mesh module, including the refractive index spatial dependency due to photoelasticity in the permittivity tensor. The derivatives of the real and imaginary parts of the optical frequency relative to the mechanical mode deformation amplitude provide – within numerical precision – an accurate calculation of the optomechanical couplings.

The moving boundary \( G_{\text{MB}} \) and photoelastic \( G_{\text{PE}} \) optomechanical couplings (in units of \( \text{Hz/m} \)) are given by (see S2 in the Supplemental Material for details):
\[
G_{\text{MB}} = \frac{\omega_0}{2} \int d\bar{r} \bar{A} \cdot \Delta \epsilon \bar{E}_m^R + \bar{E}_m^L \Delta \epsilon^{-1} \bar{E}_m^R, \tag{1a}
\]
\[
G_{\text{PE}} = \frac{\omega_{0,\beta}}{2} \int d\bar{r} \bar{E}_m^L \cdot \Delta \epsilon \bar{p} \cdot \bar{S} \bar{E}_m^R, \tag{1b}
\]
where \( \omega_0 \) is the unperturbed (complex) frequency of the optical resonator, \( \bar{A} \) is the unit-normalized mechanical displacement associated with the elastic strain \( \bar{S}, \bar{p} \) is photoelastic tensor, \( \perp \) and \( \parallel \) denote the perpendicular and parallel field components at the mechanical resonator’s surface, and \( \Delta \epsilon = \epsilon_1 - \epsilon_2, \Delta \epsilon^{-1} = (\epsilon_1)^{-1} - (\epsilon_2)^{-1} \), are related to the permittivities of the guiding \( (\epsilon_1) \) and surrounding \( (\epsilon_2) \) materials. To account for material dispersion, \( \epsilon \) is taken to be spatially and frequency dependent (see Supplemental S2). The couplings defined in Eqs. 1a, 1b are complex numbers and can be directly associated with their dispersive and dissipative components via \( G_{\omega,\text{MB/PE}} = -\Re(G_{\text{MB/PE}}) \) and \( G_{\epsilon,\text{MB/PE}} = 2\Im(G_{\text{MB/PE}}) \), where \( \Re \) and \( \Im \) denote real and imaginary parts.

The zero-point dissipative optomechanical coupling rates \( g_k = G_{\epsilon,x_{\text{zp}}} \), where \( x_{\text{zp}} \) is the zero-point fluctuation of the mechanical mode), are displayed in Fig. 2 c). Even in high-Q regimes the real modal volume approach fails to predict \( g_k \) accurately, while the discrepancy grows at smaller gaps. This analysis shows that the complex modal volume approach should be used to capture non-Hermitian features in optomechanical systems. On the
Dispersive side \((g_\omega = G_\omega x_{\text{exp}})\), shown in Fig. 2 d), striking differences in the predictions between the real and complex modal analysis arise for tighter gaps, indicating that in strongly dissipative systems, our approach should be used to correctly calculate \(g_\omega\). Our results contrast with approaches used in state-of-the-art experiments for the calculation of the optomechanical coupling, where real modal volumes are still the standard. Notice, however, that both real and complex modal volume calculations approach each other for the dispersive coupling in the high-Q regime.

**Integrated photonics resonators.** — We now turn our attention to experimental results where an appreciable dissipative coupling is observed in order to put the present formalism to test. To the best of our knowledge, the split-beam nanocavity of Ref. [18] displays the largest \(g_\omega\) reported to date in integrated devices. This 1-D photonic crystal structure is composed by two independent mechanical cantilevers separated by a gap which also serves as defect for high-Q optical confinement. The optical and mechanical modes, which will be considered here, are displayed in Fig. 3 a). Fabrication imperfections break the vertical symmetry of the system, yielding an offset (here denominated \(z\)-gap) between the two cantilevers, thus degrading the optical Q-factors as observed in Fig. 3 b). The reported uncertainty in \(z\)-gap is \(< 25\text{ nm}\) and is represented by the yellow shaded areas in Fig. 3, while experimental values for the optical quality factors are represented by the region covered by the blue strip. The Q-factors were calculated by introducing a carefully implemented PML at the boundaries of the simulation domain. The values obtained for \(z\)-gap = 25 nm \((Q_{\text{sim}} \approx 14k)\) agree within 15% to the experimental values \((Q_{\text{exp}} \approx 12k)\); such small discrepancies are possibly due to design differences between the fabricated and simulated devices.

The optomechanical couplings are finally evaluated perturbatively by using both real and complex modal volumes while accounting for PE and MB contributions. The measured dispersive coupling \(G_\omega\) (light red strip) agrees well with both perturbation series, as shown in Fig. 3 c). This is in accordance with the discussion for the ring resonator, where both formalisms accurately describe \(G_\omega\) for low optical dissipations. Results for \(G_\kappa\), are displayed in Fig. 3 d); in this example, the real and complex modal volumes yield strikingly different predictions, while agreement with measured values (cyan strip) is only obtained for the latter. This is again, well described by our toy model where the dissipative coupling is only correctly captured by the modified perturbation series, even for high-Q resonators. This analysis reinforces the physical significance of the complex modal volume and brings in a new tool for engineering non-Hermitian effects that may play a noteworthy role in optomechanics, deeply impacting optomechanical transduction.

**Plasmonic resonators.** — Molecular optomechanics, or the interplay between plasmonic resonators and molecular vibrational modes, are particularly promising systems for ultra-high optomechanical coupling. Although the phenomena of SERS or TERS — where the Raman scattering from a molecule is drastically enhanced when near a metallic structure — has been known for decades, its connection with cavity optomechanics was only pointed out recently [38, 39]. Some important features, observed in experiments, and absent in the standard theoretical treatment, were qualitatively captured by the introduction of dynamical backaction. Despite the low optical Qs found in such plasmonic structures, there are currently no reports on the role played by the dissipative coupling.

The couplings between molecular vibrational and plasmonic modes are usually calculated in a point-dipole approximation (polarizability \(\alpha(x)\)). The dipole is chosen...
FIG. 3. (a) Optical and mechanical modes of the split-beam nanocavity. (b) Optical Q-factor as a function of the offset between the two cantilevers. Inset: optical mode for 100 nm z-gap. (c) Dispersive coupling $G_{\omega}$ as a function of z-gap. (d) Dissipative coupling $G_{\kappa}$ as a function of z-gap. The yellow and light colored strips represent respectively the uncertainty in z-gap and measured values reported in [18].

to be optimally aligned with the polarization of the interacting electric field and located at $r_{\text{max}}$ where electric field amplitudes are maximum ($E_{\text{max}}^2$). Using the complex modal volume formalism, the total optomechanical coupling $G = -G_{\omega} + jG_{\kappa}/2$ reads:

$$G = -\frac{\omega_p}{2}\frac{d\alpha}{dx} \frac{E_{\text{max}}}{\int E \cdot \varepsilon E dV} = -\frac{\omega_p}{2\varepsilon(R_{\text{max}})}V_m \frac{d\alpha}{dx},$$

where $V_m$ is the complex modal volume and $\omega_p$ the plasmonic mode unperturbed frequency. In practice, $\frac{d\alpha}{dx}$ is obtained through the Raman activity/cross section of the vibrational mode under consideration.

We demonstrate the relevance of the dissipative coupling in plasmonic resonators by numerically simulating a gold NPoM scheme depicted in Fig. 4 a). The sphere (radius 70 nm) and mirror are spaced by a 1 nm wide dielectric gap ($\varepsilon/\varepsilon_0 = 2.1$). The plasmonic modes supported are assumed to interact with Biphenyl-4-thiol molecules (BPT, frequency $\Omega_m/(2\pi) = 47.52$ THz (196.5 meV), mechanical damping rate $\Gamma_m/(2\pi) = 169.3$ GHz (0.7 meV)), which are known to form self-assembled monolayers, favoring optimal optomechanical coupling. A plasmonic mode of the sphere ($Q \approx 6$, $\omega_c/(2\pi) \approx 440$ THz), consistent with bonding dimer plasmon modes reported [11] in similar structures, is used. Electromagnetic calculations were performed using a Drude-Lorentz model for the gold structures along with a PML implementation (section S3 in the Supplemental Material). We also neglect non-classical correlations due to electronic length scales [40]. The zero-point optomechanical couplings were evaluated in Fig. 4 b), displaying a strikingly strong dissipative coupling $|g_{\kappa}| > |g_{\omega}|$, which indicate pronounced non-Hermitian physics.

To understand the effect of this new physics, we consider optical cooling/amplification of a single molecule (see S4 in the supplemental material), as a function of the detuning between laser and plasmonic resonance $\Delta/\kappa$ (Fig. 4 c)), in the presence and the absence of the dissipative coupling. We used a quantum noise approach [16, 41] which considers a fixed input power from an external laser that is chosen to yield a single plasmon at resonance ($\Delta = 0$). Since such structure is often excited using a focused light beam, we can approximate $\kappa_c \ll \kappa_i$. Our results show that the presence of $g_{\kappa}$ significantly degrades optical amplification. Such effect is ultimately connected to the relative phase between $g_{\kappa}$ and $g_{\omega}$, which generates an out-of-phase modulation in the number of photons with respect to the purely dispersive case. Our calculations do indicate a blue-shift in the maximum Raman enhancement factor when compared to previous calculations which only considered $g_{\omega}$. This feature has been observed experimentally and is reported in Refs. [38, 42], indicating that dissipative optomechanical physics could be already present in state-of-the-art experiments.

Finally, we explore the experimental case reported in Ref. [11], where $N \approx 100$ BPT molecules are coupled to a single plasmonic mode, giving rise to molecular coherent collective self-sustaining oscillations, effectively enhancing the optomechanical coupling by a factor $\sqrt{N} = 10$. We choose pump laser frequencies that yield maximal amplification for the case with $(\omega_l(g_{\kappa} \neq 0))$ and without $(\omega_l(g_{\kappa} = 0))$ dissipative coupling. The driving frequencies are schematically shown at scale in Fig. 4 d1). In Fig. 4 d2) we observe that the onset of instability is verified to occur at $\approx 60\%$ higher driving powers if dissipative coupling is considered. Our analysis shows that $g_{\kappa}$ plays a major role in the molecular dynamics. Neglecting its effects may lead to inaccurate readout for $g_{\omega}$ if dynamical backaction is used as a method for its evaluation, ultimately impairing the prospect of quantum optomechanics in this platform.

**Conclusion.** — We have proposed and shown that complex modal volumes can be used to capture effects that arise from the non-Hermiticity in optomechanical systems. The formalism is underpinned by modal analysis and therefore provides insight into the engineering of such effects in integrated photonics. This may open new possibilities for dissipative coupling in ultra-high Q devices ($Q > 10^4$), where optomechanical transduction through this mechanism is enhanced. This work also points out the relevance of the phenomenon for low-Q plasmonic devices, where dynamical backaction is greatly affected by the presence of dissipation. This interplay is potentially important for a proper understanding of the Raman spectrum of molecules and for prototyping the
next generation of plasmonic devices. Lastly, the effects of such coupling in optomechanical systems operating in the PT-symmetric regime or near an exceptional point remains vastly unexplored. Since those rely on intrinsically non-Hermitian physics, the dissipative coupling may be particularly important and thus a source for a plethora of interesting phenomena.

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S1. ELECTROMAGNETIC EIGENVALUE PROBLEM

The eigenmodes of an optical system are calculated from the generalized eigenvalue equation for electromagnetism:

\[ \nabla \times \nabla \times \vec{E}(r) = \left( \frac{\omega}{c} \right)^2 \varepsilon(r) \vec{E}(r). \]  

(S1)

As pointed out in the main text, the optomechanical interaction is perturbatively calculated by introducing a change in the dielectric constant \( \Delta\varepsilon \) which is calculated from the mechanical modes profiles. However, the typical description of such phenomenon is based on very particular properties of the operators \( \nabla \times \nabla \times \) and \( \varepsilon \), namely: they are both taken as Hermitian and \( \varepsilon \) is positive semi-definite. A series of theorems in linear algebra prove that in this case, the solutions to Eq. S1 are a complete, power-orthogonal set, and its eigenvalues are real.

In the case of dissipative systems, this is not necessarily true: frequencies become complex and operators are no longer Hermitian. Specifically, we now allow a complex dielectric constant \( \varepsilon \) which violate some hypothesis of the theorems that applied earlier. Consequently, eigenfunctions are no longer power-orthogonal and, in general, do not form a complete set. From a numerical perspective, the problem of completeness is circumvented by noticing that simulation domains are finite and therefore fall in a special case, where completeness is recovered.

We tackle the problem of orthogonality by investigating the following generalized eigenproblem (EVP) and its transpose:

\[ \hat{A} \vec{\Psi}_k^R = \lambda_k^R \hat{B} \vec{\Psi}_k^R, \]  

(S2)

\[ \hat{A}^T \vec{\Psi}_k^L = \lambda_k^L \hat{B}^T \vec{\Psi}_k^L, \]  

(S3)

where \( \hat{A} \) and \( \hat{B} \) are operators, the superscript “\( T \)” denotes the transposition and \( \lambda_k^R \) (\( \lambda_k^L \)) is the \( k \)th right (left) generalized eigenvalue. The transposed EVP is sometimes called adjoint EVP (superscript \( \dagger \)) \[1\], while its eigenvectors are called left (\( \vec{\Psi}_k^L \)) eigenvectors. In the same spirit, eigenvectors of the EVP are the right (\( \vec{\Psi}_k^R \)) eigenvectors \[2, 3\]. The EVP and transposed EVP share the same spectrum. At this point we do not restrict our discussion to any specific class of operators as long as no singularities, e.g. exceptional points, are present.

We now recall the definition of an adjoint (transposed) \[4\] operator. Given a pseudo-inner product \[5\], denoted by \( <,> \) acting on a vector Hilbert space (of square-integrable functions), the transposed operator of \( \hat{\Theta} \) is such that:

\[ < \vec{\Psi}_k^L, \hat{\Theta} \vec{\Psi}_k^R >= < \hat{\Theta}^T \vec{\Psi}_k^L, \vec{\Psi}_k^R >. \]  

(S4)

Defining:

\[ < \vec{\Psi}_k^L, \vec{\Psi}_k^R >= \int dV \vec{\Psi}_k^L \cdot \vec{\Psi}_k^R, \]  

(S5)

one may show that:

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\[ \int dV \vec{\Psi}_k \cdot \hat{A} \vec{\Psi}_k = \int dV \hat{A}^T \vec{\Psi}_k \cdot \vec{\Psi}_k \implies \int dV (\lambda_k^R \vec{\Psi}_k \cdot \hat{B} \vec{\Psi}_k - \lambda_k^L \hat{B}^T \vec{\Psi}_k \cdot \vec{\Psi}_k) = 0. \] (S6)

From the above definitions, it is straightforward to prove that if the operator \( \hat{\Theta} \) is a matrix of complex numbers, the transposed operator is obtained by \( \Theta_{ij} = \Theta_{ji}^T \); while in the case of differential operators, the transpose can be found using integration by parts.

Taking into account the fact that the EVP and its transpose share the same spectrum, Eq. S6 may be simplified to:

\[ (\lambda_k^R - \lambda_k^L) \int dV \vec{\Psi}_k \cdot \hat{B} \vec{\Psi}_k = 0, \] (S7)

which is the statement of bi-orthogonality.

### A. Standing-wave resonators

The task is to find the transpose of the operators of \( \varepsilon(r) \) and \( \nabla \times \nabla \times \). This will allow us to relate left and right eigenvectors. In electromagnetism, the operator \( \hat{B} \) translates into the permittivity tensor \( \varepsilon \), which in reciprocal media obeys \( \varepsilon = \varepsilon^T \). Also, for standing-wave resonators, \( \hat{\Theta} = \nabla \times \nabla \times \) is symmetric, i.e. \( \hat{\Theta} = \hat{\Theta}^T \), as can be shown through integration by parts of this operator acting on any of the entries of Eq. S4 [6]. In other words, the EVP and transposed EVP are the same, therefore the left and right eigenvectors are the same:

\[ \vec{E}^L_{\vec{\beta}} = \vec{E}^R_{\vec{\beta}}, \] (S8)

Finally, the orthogonality relations become:

\[ \left( \frac{\omega_k^2}{c^2} - \frac{\omega_k^2}{c^2} \right) \int dV \vec{E}_{k'} \cdot \varepsilon \vec{E}_k = 0, \] (S9)

where we dropped all the superscripts and all fields are right eigenvectors of the EVP.

### B. Traveling-wave resonators

An important case in photonics regards traveling-wave resonators. If the system is periodic, the eigenmodes are Bloch-Floquet type functions:

\[ \vec{\Psi}_{\vec{\beta}}(\vec{r}) = \vec{\psi}_{\vec{\beta}}(\vec{r}) e^{i\vec{\beta} \cdot \vec{r}}, \] (S10)

where \( \vec{\psi}_{\vec{\beta}}(\vec{r}) \) fulfills the periodicity condition for a lattice parameter \( \vec{R} \), i.e. \( \vec{\psi}_{\vec{\beta}}(\vec{r} + \vec{R}) = \vec{\psi}_{\vec{\beta}}(\vec{r}) \) and \( \vec{\beta} \) is a vector of the reciprocal lattice, or, in photonics, the propagation vector. This ansatz allows us to modify the electromagnetic eigenvalue problem in terms of the propagation vector \( \vec{\beta} \) and the periodic function \( \vec{E}^R_{\beta}(\vec{r}) \):

\[ (\nabla + i\vec{\beta}) \times (\nabla + i\vec{\beta}) \times \vec{E}^R_{\beta}(\vec{r}) = \varepsilon(\vec{r}) \left( \frac{\omega_{\beta}}{c} \right)^2 \vec{E}^R_{\beta}(\vec{r}). \] (S11)

The transpose of the equation above may again be obtained by integration by parts:

\[ (\nabla - i\vec{\beta}) \times (\nabla - i\vec{\beta}) \times \vec{E}^L_{\beta}(\vec{r}) = \left( \frac{\omega_{\beta}}{c} \right)^2 \varepsilon(\vec{r}) \vec{E}^L_{\beta}(\vec{r}). \] (S12)

where it finally becomes clear that if the system is periodic, the transpose equation is equivalent to that of counter-propagating modes, therefore obtaining:

\[ \vec{E}^L_{\beta} = \vec{E}^R_{-\beta}. \] (S13)
C. Dispersive media

When we consider dispersive materials, the orthogonality relations and perturbation theory results must be modified. The wave equation for dispersive media in frequency domain reads:

\[
\nabla \times \nabla \times \vec{E}^R_\omega(\vec{r}) = \left(\frac{\omega}{c}\right)^2 \varepsilon(\vec{r}, \omega) \vec{E}^R_\omega(\vec{r}).
\]

(S14)

Considering a left eigenmode \(\vec{E}^L_\omega\), with frequency \(\omega'\), and projecting both sides of the equation above on it, one gets:

\[
\int dV \vec{E}^L_\omega(\vec{r}) \cdot \nabla \times \nabla \times \vec{E}^R_\omega(\vec{r}) = \left(\frac{\omega}{c}\right)^2 \int dV \vec{E}^L_\omega(\vec{r}) \cdot \varepsilon(\vec{r}, \omega) \vec{E}^R_\omega(\vec{r}).
\]

(S15)

Integrating the LHS by parts while neglecting surface terms, yields:

\[
\int dV \vec{E}^L_\omega(\vec{r}) \cdot \nabla \times \nabla \times \vec{E}^R_\omega(\vec{r}) = \left(\frac{\omega}{c}\right)^2 \int dV \vec{E}^L_\omega(\vec{r}) \cdot \varepsilon(\vec{r}, \omega) \vec{E}^R_\omega(\vec{r}) = 0.
\]

(S16)

This is a statement of the orthogonality relations in dispersive media. The cases for standing/traveling-wave resonators are directly obtained by replacing \(\vec{E}^L_\omega\) with respectively \(\vec{E}^R_\omega\) or \(\vec{E}^R_{\omega'}\), where in the latter, the index \(-\omega'\) indicates that the counter-propagating right eigenmode should be used in the calculations.

S2. PERTURBATION THEORY

For brevity, we state only the results for the dispersive/standing-wave case, as the non-dispersive is directly obtained from it. With the orthogonality relations derived in mind, we may come back to Eq. S14 and derive a novel perturbation series expansion. Our goal is to find an expression to the shift in frequency \(\Delta \omega\) of a given eigenmode \(\vec{E}_{\omega_0}(\vec{r})\) due to a modification in the permittivity of the system \(\Delta \varepsilon\). We make the following substitutions:

\[
\varepsilon(\vec{r}, \omega) \rightarrow \varepsilon(\vec{r}, \omega_0) + \eta \frac{\partial \varepsilon(\vec{r}, \omega)}{\partial \omega} \bigg|_{\omega_0} \Delta \omega + \eta \Delta \varepsilon(\vec{r}, \omega_0),
\]

(S17)

\[
\omega \rightarrow \omega_0 + \eta \Delta \omega,
\]

(S18)

\[
\vec{E}_\omega(\vec{r}) \rightarrow \vec{E}_{\omega_0}(\vec{r}) + \eta \Delta \vec{E}_{\omega_0}(\vec{r}),
\]

(S19)

where \(\eta\) is the perturbation parameter and \(\omega_0\) is the unperturbed frequency. Importantly, the term \(\frac{\partial \varepsilon(\vec{r}, \omega)}{\partial \omega}\) accounts for the change in permittivity due to dispersion.

Assuming that \(\Delta \vec{E}_{\omega_0}(\vec{r})\) can be expanded in the quasi-normal modes of the system, i.e. \(\Delta \vec{E}_{\omega_0}(\vec{r}) = \sum_{\omega'} c_{\omega'} \vec{E}_{\omega'}(\vec{r})\), we may project the left and right sides of equation Eq. S14 (under the rules in Eq. S19) onto the left eigenvector \(\vec{E}^L_{\omega_0}(\vec{r}) = \vec{E}_{\omega_0}(\vec{r})\). The first correction (order \(\eta\)) to the eigenvalue is given by:

\[
\Delta \omega = -\frac{\omega_0}{2} \frac{\int dV \vec{E}_{\omega_0}(\vec{r}) \cdot \Delta \varepsilon(\vec{r}) \vec{E}_{\omega_0}(\vec{r})}{\frac{2}{\omega_0} \frac{\partial \varepsilon(\vec{r}, \omega_0)}{\partial \omega} \vec{E}_{\omega_0}(\vec{r})}.
\]

(S20)

The analogous expression for the traveling-wave case is given in the main text and can be obtained by projecting Eq. S14 on the counter-propagating pair of \(\vec{E}_{\omega_0}\). An important remark must be made here: since the system is non-Hermitian, \(\omega_0\) and the volume integrals in Eq. S20 are, in general, complex numbers, and thus the imaginary part of \(\Delta \omega\) can be related to the modification in the losses of the system.

We may now proceed and derive expressions for the optomechanical coupling in microresonators, with little importance given to the hermiticity of operators and under the hypothesis of the medium’s reciprocity. In the optomechanical case \(\Delta \varepsilon(\vec{r}) \propto x\), where \(x\) is the mechanical displacement. The first order correction to the optical losses due to a change in the system’s permittivity is:
\[ \Delta \kappa_i(x) = 2 \text{Im}\{\Delta \omega(x)\}, \]  
\hspace{1cm} (S21)

where we assumed a harmonic dependency of the kind \( \exp(j\omega t) \). The rate of variation in dissipation due to deformations is:

\[ G_\kappa = 2 \frac{\partial \text{Im}\{\omega\}}{\partial x}, \]  
\hspace{1cm} (S22)

where we defined \( G_\kappa = \frac{\partial \kappa_i}{\partial x} \). On the dispersive side:

\[ G_\omega = -\frac{\partial \text{Re}\{\omega\}}{\partial x}, \]  
\hspace{1cm} (S23)

where the negative sign is adopted out of convention and is accounted for in the equations for the optical mode evolution. The zero-point couplings are defined as \( g_\omega = x_{zpf} G_\omega \) and \( g_\kappa = x_{zpf} G_\kappa \), where \( x_{zpf} = \sqrt{\hbar/(2m_{\text{eff}} \Omega_m)} \) is the zero-point fluctuation for the mechanical displacement.

### S3. ANALYTIC EXAMPLE

As mentioned in the main text, QNMs (or the eigenmodes of an open, lossy system) are known to yield divergent real mode volumes (which are based on power-orthogonality), while complex mode volumes remain finite. As shown in [7], the bi-orthogonal product displayed here is an invariant through complex coordinate stretching transforms such as in perfectly matched layers (PML), hence analytic results are expected to be robust to PML position changes. In that spirit, notice that all volume integrations described before should also be performed within the PML domain. This gives us a very practical way to compute those modal volumes numerically.

In this section, we use an analytical model to show the convergence of the bi-orthogonal product in open systems and also for insight on how to correctly handle numerical implementations of the PML and complex modal volumes. We consider an infinitely long nanocylinder, (GaAs, radius \( R = 250 \text{nm} \)), which admits whispering-gallery type traveling waves, surrounded by an absorbing layer (PML) also implemented analytically. The PML and cylinder are separated by a gap that will be varied to show the invariance of the bi-orthogonal product with respect to the PML position. For that purpose, we consider perturbations induced by boundary movement (increase in \( R \)) in the cylinder and calculate (using the moving boundary optomechanical coupling shown in the main text) the shift in frequency of the resonator.

#### A. Optical modes

The invariance in \( z \) of our system allows to uncouple the \( \hat{z} \) fields from the \( \hat{r} \) and \( \hat{\phi} \) fields, where we adopted cylindrical coordinates in Eq. S1. An arbitrary solution can be obtained by superposition of two field polarizations - \( \vec{E}_z \neq 0, \vec{H}_z = 0 \) (TM) and \( \vec{E}_z = 0, \vec{H}_z \neq 0 \) (TE). Here, we consider only TE modes.

Using the ansatz \( \vec{H}(r) = \Psi_z(r) \exp(-jm\phi) \hat{z} \), the solutions to the \( \hat{z} \) fields are given by the Bessel functions of first and second kinds:

\[ \Psi_z = AJ_m\left(\frac{n\omega c}{r}\right) + BY_m\left(\frac{n\omega c}{r}\right), \]  
\hspace{1cm} (S24)

In the case of TE modes, only the \( \hat{z} \) component of the magnetic field is non-zero, therefore the electric field may be obtained through Maxwell’s relations as:

\[ \vec{E}(\vec{r}) = \frac{j}{\omega \varepsilon} \nabla \times \vec{H}(\vec{r}). \]  
\hspace{1cm} (S25)

Counter-propagating modes (\( m \to -m \) in Eq. S24) are easily obtained from the solutions of the original problem by flipping signal of the \( \hat{r} \) (and \( \hat{z} \)) components of the electric field. This will be useful when using the pseudo-inner product defined above.
While this solution applies to all the domains (dielectric, air and PML), certain simplifications can be made: denoting $R$ as the cavity radius, for $r < R$ we may set $B = 0$, otherwise the solution would be divergent at $r = 0$. The PML is implemented analytically through the map $r \rightarrow R + \text{gap} + (1 - j)\sigma_0 \times (r - R - \text{gap})$, where $\sigma_0 = 5$ is chosen. Furthermore, we limit the domain of our solution by placing a perfect electrical conductor at radius $r = R_{\text{PEC}}$, positioned far enough from the PML/air interface such that convergence, within numerical accuracy, is achieved.

For comparison, we implement the same system in COMSOL®, although a thickness $t = 250$ nm for the cylinder was picked, as shown in Fig. S1 a2). This thickness will be important for calculating the mechanical modes supported by the structure, affecting its motional mass and therefore its frequencies. The numerical implementation mimics the invariance in $z$ through the usage of appropriate boundary conditions, e.g. the top and bottom of the nanocylinder are taken to be perfect magnetic conductors. Note that this choice filters out TM modes of the solutions and is therefore consistent with our TE modal analysis. The optical mode profile, in the presence of the PML, is displayed in Fig. S1 b), for the case of gap = 1.7 µm, obtained through numerical (upper) and analytic (lower) calculations.

### B. Mechanical modes

The eigenvalue equation for the acoustic normal modes of the structure is given by:

$$ -\rho \Omega_m^2 \vec{U}(\vec{r}) = \nabla \cdot \mathbf{T}, $$

where $\rho$ is the mass density of the material, $\vec{U}(\vec{r})$ is the mechanical mode profile and $\mathbf{T}$ is the stress tensor, which can be found by contraction between the stiffness ($c$) and strain ($S$) tensors. We again use the invariance of the system and thus use an ansatz $\vec{U}(\vec{r}) = U(r)\hat{r}$, which will capture purely mechanical breathing modes of the structure. This choice yields only two non-vanishing terms in the strain tensor, namely $S_1 = \frac{dU}{dr}$ and $S_2 = \frac{U}{r}$ (in Voigt notation), therefore the solutions to Eq. S26 are:

$$ U(r) = AJ_1 \left( \frac{\Omega_m r}{v_L} \right) + BY_1 \left( \frac{\Omega_m r}{v_L} \right), $$

where $v_L = \sqrt{\frac{c_{11}}{\rho}}$. Physical solutions must have $B = 0$ while $A$ is chosen based on normalization requirements. The mechanical frequency $\Omega_m$ is found by imposing free boundary-conditions ($\mathbf{T} \cdot \hat{r} = 0$), yielding the following transcendental equation:

$$ \frac{c_{11}}{2v_L} \left[ J_0 \left( \frac{\Omega_m R}{v_L} \right) - J_2 \left( \frac{\Omega_m R}{v_L} \right) \right] + \frac{c_{12}}{\Omega_m R} J_1 \left( \frac{\Omega_m R}{v_L} \right) = 0, $$

which can be self-consistently solved.
FIG. S2. a) Optomechanical couplings $g_\omega$, $g_\kappa$, and b) (real) frequency $\omega$ and optical linewidth $\kappa$ as a function of the gap size for an infinite nanocylinder. Both simulated and analytic calculations are shown, the latter matches the averaged simulated values. c) and d) display the same analysis for the NPoM in the main text.

Numerically, the mechanical mode is computed by considering symmetry boundary conditions ($\hat{n} \cdot \vec{U} = 0$) on the top and bottom of the nanocylinder, imposing the absence of any mechanical deformations in the $z$ direction, while the $\phi$ dependency is accounted for by choosing the azimuthal number $m = 0$. The values found for the mechanical frequencies are $\Omega_m = 6.418$ GHz (numerical) and $\Omega_m = 6.417$ GHz (analytic), displaying excellent mutual agreement.

C. Domain of validity

We finally explore the extent of validity of the bi-orthogonal perturbation theory. Our aim here is to demonstrate the accuracy and also show how large a deformation has to be (relatively to the size of the structure), in order to significantly deviate from the first order perturbation theory expansion. We consider only boundary movement (increase in $R$), which allows for completely analytical results, in contrast to photoelasticity, where the non-trivial dielectric function imposed by the mechanical strain renders an electromagnetic eigenvalue equation that can only be solved numerically. Also, an increase in $R$ is exactly emulated by the mechanical breathing mode (since displacements are purely radial, as shown in the inset of Fig. S1 c)), yielding direct comparison between exact and perturbative analytic calculations.

We compute the frequency shift as a function of the deformation of the nanocylinder boundary. As shown in Fig. S1 c), the exact and perturbative calculated shifts are in good agreement up to displacements of about 10 nm, about 4% of the nanocylinder radius. This puts optomechanical calculations safely within the validity region, where displacements are typically $\ll 0.1\%$ of the device’s relevant size.

D. Invariance of the bi-orthogonal product with the PML position

We now turn our attention to investigating the invariance of the integral in Eq. S16 with respect to the size of the air domain. This will give us insight on how to correctly position the PML in our numerical study. We start by comparing the results for the zero-point optomechanical couplings in the numerical and analytic models for the nanocylinder. We sweep over the gap distance between the dielectric’s and PML boundaries. For each gap, the optical
modes, \(g_e\) and \(g_m\) are evaluated. The result is displayed in Fig. S2 a) where simulated points are spline-interpolated as a guide for the eye. Oscillations in the simulation results are observed, while constant values are obtained in the analytic calculations. The oscillations in \(g_e\) (\(g_m\)) display the same periodicity of the imaginary (real) parts of \(\vec{E}_\omega\) \(\vec{E}_z\), hence the \(\frac{\pi}{2}\) phase difference observed between the blue and red curves. The increasing amplitude is related to the exponential growth of the QNM fields as a function of distance. The oscillatory behavior is only present due to the finite-element method discretization in the PML domain, invariably leading to reflection of incoming waves, which in turn, deeply affects the phase information (ubiquitous and conserved in the bi-orthogonal formalism, since no pairs of complex conjugate fields appear) of the calculated modes. Averaging over many oscillations, however, leads to correct values for both dissipative and dispersive couplings, as shown by the light shades of blue and red, optimally overlapping with analytically obtained values. Mesh refinement within the computational power available was not observed to yield significantly better results than those displayed in here.

The same features are present if the frequency and linewidth of the optical modes are considered. Results are displayed in Fig. S2 b) and notably, the relative error in this case (amplitude/average ratio) is much smaller than in the previous case. This analysis indicates that while Q-factors are typically used as benchmark in typical implementations of PMLs, if complex modal volumes are to be considered, further care is needed.

Lastly, we show in Figs. S2 c) and d) an analogous treatment in the case of the nanoparticle-on-a-mirror scheme presented in the main text. Here, the gap length is taken to be the distance between the radius of the nanosphere and the PML boundary. The oscillations in the simulated values are again verified and averaging was used for all relevant calculations.

**S4. HAMILTONIAN FORMALISM AND DYNAMICAL BACKACTION**

The Hamiltonian interaction of the optomechanical system with both dissipative and dispersive interaction is given by: \(\hat{H} = \hbar \omega_c \hat{a}^\dagger \hat{a} + \hbar \omega_m \hat{b}^\dagger \hat{b} + \hat{H}_\kappa + \hat{H}_{\Gamma_n} + \hat{H}_{\text{OM}}\) with:

\[
\hat{H}_{\text{OM}} = - \left[ \hbar \tilde{g}_{\omega} \hat{a}^\dagger \hat{a} + i \sqrt{\frac{\kappa}{2\pi\rho}} \frac{\hbar \tilde{g}_\kappa}{2\kappa} \sum_q (\hat{a}^\dagger \hat{c}_q - \hat{c}_q^\dagger \hat{a}) \right] \hat{x},
\]
where \(\hat{a}(\hat{a}^\dagger)\) and \(\hat{c}_q(\hat{c}_q^\dagger)\) are the bosonic annihilation (creation) operators for the cavity and bath optical modes respectively, \(\kappa = \kappa_e + \kappa_i\) is the total cavity decay rate. \(\hat{x}\) is the mechanical position operator, and \(\rho\) denotes the density of states of the optical bath, treated as a constant for the relevant frequencies. Equation S29, along with Hamiltonians for the isolated optical/acoustic modes and their respective damping arising from interactions with the environment, allow us to formulate the dynamics of the system at the level of quantum Langevin equations \([8, 9]\). In the NPoM case exemplified in the main text, we may approximate \(\kappa \approx \kappa_i(x)\), leading to the force spectrum \((S_{FF}(\Omega))\) given by \([9]\):

\[
S_{FF}(\Omega) \approx \frac{\hbar g_e^2}{4\kappa x_{zpf}^2} n_c (\Delta + \Omega + 2\kappa (g_m/g_e))^2 + (\kappa/2)^2 + (\Delta/2)^2 + (\kappa/2)^2 \approx \frac{1}{16} \left[ (\Delta - \Omega_m + 2\kappa (g_m/g_e))^2 + (\kappa/2)^2 \right].
\]

where \(n_c\) is the number of photons inside the cavity, and \(\Delta = \omega_i - \omega_c\) is the detuning between the laser drive \((\omega_i)\) and cavity \((\omega_c)\) frequencies. Fermis Golden Rule for transitions between states with \(n\) and \(n \pm 1\) phonons results in an optically induced mechanical damping \(\Gamma_{\text{OM}} = \Gamma_\downarrow - \Gamma_{\uparrow}\) \((\Gamma_{\uparrow(\downarrow)} = \frac{\kappa}{2\kappa} S_{FF}(\mp \Omega_m))\) given by:

\[
\Gamma_{\text{OM}}(\Delta) \approx \frac{\kappa}{4 \kappa} \sum_{\Omega_m} \left((\Delta + \Omega_m + 2\kappa (g_m/g_e))^2 + (\kappa/2)^2 \right) + \frac{(\Delta - \Omega_m + 2\kappa (g_m/g_e))^2 + (\kappa/2)^2}{(\Delta - \Omega_m)^2 + (\kappa/2)^2}.
\]

In order to gain some insight we evaluate Eq. S31 in the good cavity limit \((\Omega_m >> \kappa)\) when driving the system at the cavity blue side \((\Delta = \Omega_m)\)

\[
\Gamma_{\text{OM}} \approx \frac{4g_e^2 n_c}{\kappa} \left[ 1 - \frac{1}{8} \left( \frac{\kappa}{\Omega_m} \right) \left( \frac{g_e}{g_m} + \frac{1}{2} \right) - \frac{1}{256} \left( \frac{\kappa}{\Omega_m} \right)^2 \left( \frac{g_e}{g_m} \right)^2 \right]
\]

where the first term gives the conventional dispersive contribution to the mechanical linewidth and the remaining terms are related to dissipative coupling. Using the values for the NPoM in the main text, we can estimate and
compare the number of photons for the self-sustained oscillation threshold ($\Gamma_{OM} = -\Gamma_m$) with ($g_\kappa \neq 0$) and without ($g_\kappa = 0$) dissipative coupling to be:

\[
\frac{n_{(g_\kappa \neq 0)}}{n_{(g_\kappa = 0)}} \approx 1 - \frac{1}{8} \left( \frac{\kappa}{\Omega_m} \right) \left( \frac{g_\kappa}{g_\omega} + \frac{1}{2} \right) - \frac{1}{256} \left( \frac{\kappa}{\Omega_m} \right)^2 \left( \frac{g_\kappa}{g_\omega} \right)^2 = 0.58, \tag{S33}
\]

which is in good agreement with the ratio of on resonance plasmon population shown in the main text, evaluated using the full and optimal detunings for Eq. S31.

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