**T-Operator Limits on Electromagnetic Scattering:**

*Bounds on Extinguished, Absorbed, and Scattered Power from Arbitrary Sources*

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We present a scheme for obtaining physical bounds on any electromagnetic scattering problem that can be framed as a net emission, scattering or absorption process. The method requires only a high level description of the design problem—the material the device will be made of, a boundary for the volume the device may occupy, and a description of the incident field or excitation current source—to simultaneously consider all structuring possibilities. Both surprising and anticipated characteristics related to material and geometric properties are observed. For the canonical case of a propagating plane wave interacting with an arbitrarily designable object contained in a spherical bounding domain of radius \(R\), the scattering cross sections in the small \(R/\lambda \ll 1\) (quasi-static) limit display a material dependence on the electric susceptibility \(\chi\) for metals corresponding to a diluted (homogenized) material response \(\propto -\text{Re}(\chi_{\text{res}})/\text{Im}(\chi)\) achieved via nanostructuring, such that for the same material loss, the performance of strong metals (\(\text{Re}(\chi) \ll -2\)) is found to be significantly weaker than that of resonant spherical nanoparticles satisfying the localized plasmon-polariton condition, \(\text{Re}(\chi_{\text{res}}) = -3\). For large radii \(R \to \infty\), achievable scattering interactions asymptote to the geometric cross section of the ball, as predicted by ray optics. Bounds on the maximum power radiated by a dipole located a distance \(d\) above the spherical bounding domain are also investigated and found to be reduced compared to prior analyses, with strong metals and dielectrics exhibiting the expected \(\propto d^{-3}\) asymptotic in the deep near field regime \(d \ll R\) but a more muted dependence on \(\text{Re}(\chi)\). The basis of the proposed method rests entirely on the applicability of scattering theory, and can thus likely be applied to acoustics, quantum mechanics, and other wave physics.

Much of the continuing appeal and challenge of linear electromagnetics stems from the same root cause: given a desired objective (enhancing radiation from a quantum emitter\(^1\)\(^5\), the field intensity in a photovoltaic\(^6\)\(^8\), the radiative cross-section of an antenna\(^9\)\(^11\), etc.) subject to practical constraints (material compatibility\(^12\)\(^–\)\(^14\), fabrication tolerances\(^15\)\(^–\)\(^17\), or device size\(^18\)\(^–\)\(^20\), there is currently no method for finding uniquely best solutions. The difficulties associated with this problem are well known\(^21\)\(^–\)\(^23\). From plasmonic resonators\(^24\)\(^–\)\(^26\) to periodic lattices\(^27\)\(^–\)\(^29\), myriad combinations of material and geometry can be employed to manipulate light, confining and thereby enhancing its interactions with matter\(^30\)\(^–\)\(^32\) or altering its characteristics\(^33\)\(^–\)\(^35\), to extraordinary but similar effect\(^36\)\(^–\)\(^37\). The wave nature of Maxwell’s equations and non-convexity of optimizations with respect to variations in material susceptibility make it challenging to discern globally optimal solutions\(^38\)\(^–\)\(^40\), often yielding designs that are sensitive to minute structural alterations\(^41\)\(^–\)\(^42\). Yet, despite these apparent challenges, computational (inverse) design techniques based on local gradient information have proven to be impressively useful\(^43\)\(^–\)\(^45\), offering substantial improvements for applications such as on-chip optical routing\(^45\)\(^–\)\(^47\), metamaterials\(^48\)\(^–\)\(^50\), nonlinear frequency conversion\(^51\)\(^–\)\(^52\), and engineered bandgaps\(^53\)\(^–\)\(^54\). The widespread success of these techniques, and their increasing prevalence, raises at least two pertinent lines of inquiry. Namely, how far can this advancement continue, and, if salient limits do exist, can this information be leveraged to facilitate design? Absent benchmarks of what is possible, precise evaluation of the merits of inverse algorithms is difficult. Failure to meet some desired level of performance may be caused by issues in the choice of objective, formulation, or parametrization.

Prior efforts to elucidate such electromagnetic performance asymptotics, surveyed briefly in Sec.\(^1\)\(^1\) have provided significant insights into a diverse collection of topics, including antennas\(^9\)\(^–\)\(^10\)\(^,\)\(^55\), light trapping\(^6\)\(^–\)\(^8\)\(^,\)\(^60\), and optoelectronic\(^61\)\(^–\)\(^64\) devices, resulting in novel contemporary design tools for specialized systems\(^39\)\(^–\)\(^65\)\(^–\)\(^67\). Yet, their domain of practical applicability is patchwork. Barring recent computational bounds established by Angeris, Vučković and Boyd\(^39\), which are limits of a different sort, and the efforts of Gustafsson et al.\(^68\) to transfer established techniques from antenna theory to nanophotonic settings\(^69\)\(^–\)\(^72\), published during the final preparation of our article and independently converging on a similar program, applicability is highly context dependent. Relevant arguments shift with circumstance\(^73\)\(^–\)\(^74\), and even within the same setting, widely recognized attributes (e.g., the impact of metallic versus dielectric response, finite object sizes, or minimum feature sizes) are often unaccounted for, leading to loose (unphysical) predictions.

In this article, we present a framework based on LaPlace duality and the scattering \(T\) operator for computing bounds on any electromagnetic objective that can be framed as a total emission, scattering, or absorption process, encompassing nearly every application mentioned above. The scheme, capturing all possible structures as well as the fundamental wave limitations contained in Maxwell’s equations, can be applied provided only a coarse description of the design problem. Specifically, only three
pieces of information are required: the material the device will be made of, the volume it can possibly occupy, and the source that it will interact with. The introduction of additional modal constraints compared to other approaches [68, 70, 75], including our recent work [78, 76], makes this information sufficient to encode the effects of the electric field susceptibility ($\chi$) and finite sizes on a per-channel basis, quantifying achievable “optical channel capacity” beyond the previously identified inverse material-resistivity ($|\chi|^2/\text{Im} \chi$) [77] and radiative efficacy [78, 76, 78] figures of merit. The limits revealed by enforcing these realities are found to be many orders of magnitude tighter than prior bounds in several settings of special relevance to photonic devices.

Two quintessential examples, selected for their generality, practicality, and openness to semi-analytic investigation are considered: planewave and dipole fields impinging on a structure contained in a ball. For the planewave case, we find that for small radii (quasi-statics), the $\chi$ dependent enhancement of the bound is only slightly larger than what is predicted by Rayleigh scattering [79]. In particular, if the chosen material does not match the resonance (localized plasmon-polariton) condition of a nanoparticle [80], the possibility of nanostructuring to attain either an increased or reduced effective medium response [81, 82] is found to offer less substantial performance benefits in this regime compared to the per-volume response of the particle, which scales with the material figure of merit $|\chi|^2/\text{Im} \chi$ [77]. More varied response and enhancement factors are encountered for larger radii as additional radiative channels and resonances emerge, with all bounds asymptoting to the area scaling consistent with ray optics in the (macroscopic) limit of large radii. In general, bounds for dielectric media begin to match what is observed for unrealistically low-loss metals once the effective size of the domain (the product of $\sqrt{\text{Re} |\chi|}$ and the domain radius) reaches the wavelength scale. Limits on the radiative power which may be extracted from a dipole source in the vicinity of the spherical domain, the radiative local density of states (LDOS), are also shown to be tighter and exhibit reduced dependence on $\chi$ compared to prior analyses [68, 83].

The totality of these findings shed light on a range of fundamental questions in electromagnetism, such as the degree of (radiative) Purcell enhancement which can be achieved through modification of the electromagnetic environment (nanostructuring), limitations on light extraction and trapping efficiency, and the relative merits of metals versus dielectrics in different settings [64, 84, 85]. Further, they also provide a substantially more quantitative outlook on which aspects of a design problem are most critical to achieving a desired level of performance. Depending on the available volume and quantity being bounded, either dielectrics or metals may be preferred. The possible gains resulting from structural optimization may or may not make a substantial difference compared to known designs. We foresee application and extensions of this framework to embedded sources and extended geometries as providing a means of formalizing, comparing, and contrasting different approaches within photonics, revealing limitations and tradeoffs among existing design paradigms in a number of technologically prescient areas (e.g. enhancing the radiative efficacy of quantum emitters [86–88], high quality factor cavities [89–91], metasurfaces optics [92, 94], cloaking [95, 97], and augmenting luminescence [98, 99] and fluorescence [100, 101]).

The article is divided into four principle sections. Sec. I begins with an overview of the $T$ operator relations governing absorption, scattering and radiative processes, followed by a statement of the wave constraints and relaxation relevant to the article. Using these tools, the calculations of limits is then cast in the language of optimization theory, and a solution in terms of Lagrangian dual is given. Sec. II explores similarities and differences of our approach with prior art. Next, in Sec. III the mechanics of a toy optimization problem are examined for an analytically soluble single channel reduction. Finally, Sec. IV provides sample bounds in the two situations described above. We remark that although only single-frequencies examples are given, extensions of this formalism to encompass broadband width objectives should present no major hurdles [63, 102]. Further, since the approach relies only on the validity and relations of scattering theory, it is likely that counterparts of all presented results exist in acoustics, quantum mechanics, and any other wave physics.

I. FORMALISM

The key to our approach for obtaining scattering bounds rests on the use of partial relaxations [103]. Past electromagnetic limits have been predominately formulated by
placing bounds on the constituent physical quantities entering an objective, and then deducing a total bound by composing the individual limits [104, 105]. We begin, alternatively, with the total relations that any physical system must satisfy, derive consequences of these relations (e.g., energy conservation) and then suppose a subset of these derived equations as algebraic constraints on an otherwise abstract optimization problem. In the absence of any constraints, a loose (possibly infinite) bound is discernible nearly by inspection; if all physical relations are respected, the difficulty of discovering a bound is likely close to finding a best inverse design solution. The crux of the matter is thus to choose constraints that retain as much essential physics as possible (as measured by agreement with known asymptotics, plausible dependencies on material response and bounding geometry, etc.) without the resulting optimization problem becoming intractable. This general procedure is detailed below.

### A. Power Objectives

Considerations of power transfer in electromagnetics typically belong to one of two categories: initial flux problems, wherein power is drawn from an incident electromagnetic field, and initial source problems, wherein power is drawn from a predefined current excitation. Initial flux problems are typical in scattering theory, and as such, our nomenclature follows essentially from this area [106]. Namely, we will denote the initial (incident, given, or bare) field with an $i$ superscript (either $\mathbf{E}^i$ or $\mathbf{J}^i$) and the total (or dressed) fields with a $t$ superscript. For a pair of initial and total quantities referring to the same underlying field, the scattered field, $s$ superscript, is defined as the difference $|\mathbf{F}^s| = |\mathbf{F}^t| - |\mathbf{F}^i|$. There is a certain appeal to transforming one of these two classes of problem into the other via equivalent fields. However, due to the additional back-interactions that can occur in initial source problems, in our experience a unified framework promotes logical slips. For this reason, the total polarization field of an initial flux problem (or total electromagnetic field of an initial source problem) will be referred to as a generated field, $g$ superscript. With this notation, scattering theory for initial flux and source problems consists of the following relations.

\[
|\mathbf{F}^s| = \frac{ik_o}{Z} \mathbf{V} |\mathbf{E}^i| , \quad |\mathbf{E}^i| = \mathbf{V}^{-1} T |\mathbf{E}^i| \\
|\mathbf{E}^t| = |\mathbf{E}^i| + \frac{iZ}{k_o} G^0 |\mathbf{F}^s| , \quad |\mathbf{E}^s| = \frac{iZ}{k_o} G^0 |\mathbf{F}^s| \\
|\mathbf{F}^g| = \frac{iZ}{k_o} G^0 |\mathbf{F}^i| , \quad |\mathbf{F}^i| = \mathbf{V} T^{-1} |\mathbf{F}^i| \\
|\mathbf{F}^t| = |\mathbf{F}^i| - \frac{ik_o}{Z} \mathbf{V} |\mathbf{E}^g| , \quad |\mathbf{E}^f| = -\frac{ik_o}{Z} \mathbf{V} |\mathbf{E}^g| \tag{1}
\]

Here and throughout, $G^0$ marks the background or environmental Green’s function, which may or may not be vacuum. The $V$ operator refers to the scattering potential (susceptibility) relative to this background (whatever material was not included when $G^0$ was computed), and $|\mathbf{E}^i|$ and $|\mathbf{J}^i|$ are similarly defined as initial fields in the background. The remaining quantities in (1) and (2) are the impedance of free space $Z$, the wavenumber $k_o = 2\pi/\lambda$ (with $\lambda$ the wavelength), and the $T$ operator, defined by the formal equality $T = (\mathbf{V} - G^0)^{-1}$ [73].

The three primary operator forms for energy transfer in an initial flux problem are the extracted power,

\[
P_{\text{flx}}^\text{ext} = \frac{1}{2} \Re [|\mathbf{E}^f|^2] = \frac{k_o}{2Z} \text{Tr} [S_E \text{Asym}[T]], \tag{3}
\]

the absorbed power,

\[
P_{\text{flx}}^\text{abs} = \frac{1}{2} \Re [|\mathbf{E}^f|^2] = \frac{k_o}{2Z} \text{Tr} [S_E (T^t \text{Asym}[V^{-1}] T)], \\
= \frac{k_o}{2Z} \text{Tr} [S_E \text{Asym}[T] - T^t \text{Asym}[G^0] T], \tag{4}
\]

and the scattered power,

\[
P_{\text{flx}}^\text{scat} = -\frac{1}{2} \Re [|\mathbf{E}^f|^2] = \frac{k_o}{2Z} \text{Tr} [S_E T^t \text{Asym}[G^0] T] \\
= \frac{k_o}{2Z} \text{Tr} [S_E \text{Asym}[T] - T^t \text{Asym}[V^{-1}] T]]; \tag{5}
\]

with $S_E = |\mathbf{E}^f\rangle \langle \mathbf{E}^f|$ denoting projection of the corresponding operators onto the incident fields. Reciprocally, the three principal forms characterizing power flow from an initial current excitation are the extracted power,

\[
P_{\text{src}}^\text{ext} = -\frac{1}{2} \Re [|\mathbf{J}^f|^2] = \frac{Z}{2k_o} \text{Tr} [S_J (\text{Asym}[G^0] + \text{Asym}[G^0 T G^0])] \\
= \frac{Z}{2k_o} \text{Tr} [S_J \text{Asym}[V^{-1}] + \text{Asym}[V^{-1} \mathbf{V} T^{-1}]], \tag{6}
\]

the radiated power,

\[
P_{\text{rad}}^\text{scat} = -\frac{1}{2} \Re [|\mathbf{J}^f|^2] = \frac{Z}{2k_o} \text{Tr} [S_J (V^{-1} T^t \text{Asym}[G^0] T V^{-1})] \\
= \frac{Z}{2k_o} \text{Tr} [S_J V^{-1} (\text{Asym}[T] - T^t \text{Asym}[V^{-1}] T) V^{-1}] \\
= \frac{Z}{2k_o} \text{Tr} [S_J \text{Asym}[G^0] + \text{Asym}[G^0 T G^0]] - \text{Tr} [S_J G^0 T^t \text{Asym}[V^{-1}] T G^0], \tag{7}
\]

and the material (loss) power,

\[
P_{\text{src}}^\text{nat} = \frac{1}{2} \Re [|\mathbf{J}^f|^2] = \frac{Z}{2k_o} \text{Tr} [S_J (G^0 T^t \text{Asym}[V^{-1}] T G^0)] \\
= \frac{Z}{2k_o} \text{Tr} [S_J (G^0 \text{Asym}[T] G^0 - G^0 T^t \text{Asym}[G^0] T G^0)], \tag{8}
\]

with $S_J = |\mathbf{J}^f\rangle \langle \mathbf{J}^f|$ denoting projection of the corresponding operators onto the initial current sources. The naming of the second and third forms, which appear less frequently than the other four, follows from the observation
that once the total source \( |I^*\) is determined the corre-
sponding electromagnetic field is generated exclusively via
the background Green's function. Hence, the energy trans-
fer dynamics of a total source are exactly those of a spe-
cial "free" current distribution. Because the only pathway
for power to flow from a current source in free space (or
lossless background) is radiative emission, \( P_{\text{rad}}\) must be in-
terpreted in this way—energy transfer into the source free
solutions of the background—and similarly, \( P_{\text{sc}}\) must be
equated with loss into the scatterer. This reversal of forms
and physics (compared absorption and scattering) is sen-
sible from the perspective of field conversion. Absorption
is the conversion of an electromagnetic field into a current,
and radiative emission the conversion of a current into an
electromagnetic field. Scattering, conversely, in an initial
flux setting is the creation of a new field of the same type,
as is material loss in an initial source setting.

Note, however, that there is a caveat to this interpre-
tation. As \( \text{Asym}[G^0] \) describes power flow into the en-
tire electromagnetic background, if the environment for
which \( G^0 \) is determined contains absorptive material then
\( \text{Asym}[G^0] \) will not correspond to actual radiation. Im-
plied meaning can be restored by appropriately altering
\( \text{Asym}[G^0] \) and using the first forms given for the radiated
powers; but, as this point will be treated in an upcom-
ing work, for the moment we will simply accept it as a limita-
tion for our study.

Setting this possibility aside, the equivalence of \( 7 \) with
radiative emission is also supported both by the analogy
between its operator form and that of the scattered power,
and by direct calculation for thermal (randomly fluctuat-
ing) currents \( 107 \). By the fluctuation–dissipation theorem
\( \langle |I^*| \rangle |I^* \rangle_{\text{ther}} = 4k_0 \Pi(\omega, T) \text{Asym}[V^{-1}] / (\pi Z) \), and so
\[
P_{\text{ther}} = -\frac{Z}{2k_0} \left( \text{Im} \left[ \langle |I^*| \rangle |I^* \rangle_{\text{ther}} \langle V^{-1} \rangle |G^0|^2 T V^{-1} \right] \right)_{\text{ther}}
\]
\[
= -\frac{Z}{2k_0} \left( \text{Im} \left[ \text{Tr} \left[ \langle |I^*| \rangle |I^* \rangle_{\text{ther}} V^{-1} T^0 G^0 T V^{-1} \right] \right] \right)
\]
\[
= \frac{2 \Pi(\omega, T)}{\pi} \text{Tr} \left[ \text{Asym}[\text{Asym}[V^{-1}] T^0 G^0 T] \right]
\]
\[
= \frac{2 \Pi(\omega, T)}{\pi} \text{Tr} \left[ \text{Asym}[T - T \text{Asym}[G^0] T^0] \text{Asym}[G^0] \right].
\]
The final line above is precisely what we have derived in
Ref. \( 74 \) from the perspective of incident radiation.

B. Scattering Constraints

As supported by \( 1 \) and \( 2 \), all scattering processes are
described by some combination of the operators \( G^0 \), \( V \) and
\( T \). Supposing that \( G^0 \) and \( V \) are determined, a defining
relation for \( T \) is thus abstractly equivalent to a solution. That
is, like Maxwell's equations, any facet of scattering theory,
beyond the properties of \( G^0 \) and \( V \), must be derivable from the
definition of the \( T \) operator \( 74 \) \( 106 \)
\[ I = [V^{-1} - G^0] T. \]

The same is true of the related \( W \) operator
\[
I = W(1 - V_{ss} G^0) = (1 - V_{ss} G^0) W,
\]
\[
W = \left[ \begin{array}{cc}
1_{bb} & 0_{bb} \\
T_{ss} G^0 & T_{ss} V_{ss}^{-1} \\
\end{array} \right],
\]
with \( b \) and \( s \) subscripts explicitly marking the domain and
codomain of each operator as either the background \( (b) \) or
scatterer \( (s) \). In complement with \( T \), \( W \) is stated to produce a
total current from an initial current \( 22 \) \( 107 \) and thus to-
gether with \( G^0 \) and \( V \) also gives a complete description of
any linear scattering process. However, in contrast to \( T \), \( W \) is
globally defined without the need to carry out a limiting
procedure at spatial locations where the scattering poten-
tial is zero \( (\gamma \to 0) \). Hence, it is more transparent when con-
currently analyzing volumes inside and outside a scatterer.

Multiplying \( 10 \) by \( T \) and forming symmetric and anti-
symmetric pairs leads to the following relations:
\[
\text{Asym}[T_{ss}] = \sum_i T_{ss} \text{Sym}[U_{fi}] T_{ss}, \quad \text{(12)}
\]
\[
\text{Sym}[T_{ss}] = \sum_i T_{ss} \text{Asym}[U_{fi}] T_{ss}, \quad \text{(13)}
\]
In these expressions, \( U = V_{ss}^{-1} - G_{0ss} \) and \( f_i \) denotes the
family of indices coupled together by \( U_{fi} \). In some com-
plete basis \( \{G_{f_i,s}\} \) for the domain. Due to the later con-
nection of each \( f_i \) with a unique radiation mode, and rela-
tions with existing literature \( 74 \) \( 73 \) \( 76 \) \( 104 \) \( 108 \) we will inter-
changeably refer to families as channels. (In other words, \( \ell \)
 denotes the \( \ell \)th block of \( U \) in the matrix representa-
tion \( \{G_{f_i,s}\} \cup \{G_{f_i,s}\} \). This definition of \( U \) implies that \( \text{Asym}[U] \)
is positive-definite.

As recently described in Refs. \( 68 \) \( 73 \) \( 76 \) \( 12 \) and
\( 13 \) have been previously shown to contain a surprising
amount of physics. Taken together, these relations give a
full algebraic characterization of power conservation \( 68 \)
\( 109 \), with \( 12 \) representing the conservation of real power
and \( 13 \) the conservation of reactive power. (The bilinear
piece of \( 13 \) is the difference of magnetic and electric en-
ergies \( 106 \).) Because both real and imaginary parts are
thus captured, when both constraints are employed there
are global requirements that must be satisfied on both the
magnitude and phase of any potential resonances.

C. Relaxations and Optimization

For the single-source problems of concern to this article,
its simplest to work with the forms described above from
the perspective of the image field resulting from the action
of \( T_{ss} \) on a given source \( \{S^{(1)}\} \), \( T_{ss} S^{(1)} \to \{T\} \). A bound
in this setting amounts to a global maximization of one of
the six power-transfer objectives, \( 3 \) – \( 8 \), taking \( \{T\} \) and a
known linear functional \( \{S^{(2)}\} \) as arguments, subject to sat-
faction of \( 12 \) – \( 13 \) as applied to the source and its image.
So long as the known fields are not altered at previously in-
cluded locations by expanding the domain, this procedure
leads to domain-monotonic growth: if \( S^{(i)} \) and \( T \) satisfy all constraints on some subdomain, then these same vectors will also satisfy the constraints if they are embedded into a larger domain. And because the value of any power objective is similarly unaffected by inclusion, the global maximum of a larger domain will always be larger than the global maximum of a smaller domain.

The above view also underlies the central relaxation, persisting throughout the remainder of the article, that makes global optimization over all structuring alternatives possible. For any true \( T \) operator, nonzero polarization currents may exist only at spatial points lying within the object. This fact will never be strictly enforced on the image of the source resulting from the action of \( T \), alleviating the need for a geometric description of the scatterer. Rather, \( |T| \) will be considered simply as an unknown vector field confined to the domain. Hence, when a bound is found, it must necessarily apply to any possible structure that can be contained in the given region, as this freedom has already been explored by the optimization. For instance, through this relaxation of structural information and the monotonicity property, a bound for a cuboid is both a bound for any device that could fit inside the cuboid (no matter how exotic), and bound for any subdomain of the cuboid (be it a sphere, needle, bounded fractal, etc.).

With this in mind, scattering operator bounds are equated with an optimization problem on \(|T|\) and \(|R|\):

\[
\begin{align*}
\max \mathcal{O} = \sum_{f_i} \text{Im} \left[ \left( S^{(i)}_{f_i} | T_{f_i} \right) \right] - \left( T_{f_i} \big| \mathcal{O} \big| T_{f_i} \right) \\
\text{such that} \quad \mathcal{G}_\mathcal{O} = \sum_{f_i} \text{Im} \left[ \left( S^{(i)}_{f_i} | T_{f_i} \right) \right] - \left( T_{f_i} \big| \text{Asym} \left[ U_{f_i} \right] \big| T_{f_i} \right) = 0, \\
\mathcal{G}_\varepsilon = \sum_{f_i} \text{Re} \left[ \left( S^{(i)}_{f_i} | T_{f_i} \right) \right] - \left( T_{f_i} \big| \text{Sym} \left[ U_{f_i} \right] \big| T_{f_i} \right) = 0,
\end{align*}
\]

(14)

The corresponding Lagrangian is given by

\[
\begin{align*}
\mathcal{L} = \sum_{f_i} \text{Im} \left[ \left( S^{(i)}_{f_i} | T_{f_i} \right) \right] - \left( T_{f_i} \big| \mathcal{O} \big| T_{f_i} \right) + \\
\zeta \left( \text{Im} \left[ \left( S^{(i)}_{f_i} | T_{f_i} \right) \right] - \left( T_{f_i} \big| \text{Sym} \left[ U_{f_i} \right] \big| T_{f_i} \right) \right) + \\
\gamma \left( \text{Re} \left[ \left( S^{(i)}_{f_i} | T_{f_i} \right) \right] - \left( T_{f_i} \big| \text{Asym} \left[ U_{f_i} \right] \big| T_{f_i} \right) \right)
\end{align*}
\]

As before, \( f_i \) denotes the \( f \)th family of basis components coupled by \( U_{f_i} = \mathcal{V}^{-1}_{f_i} - \mathcal{G}_0 \). The constraints \( \mathcal{G}_\mathcal{O}, \mathcal{G}_\varepsilon \) and \( \mathcal{G}_\gamma \) are determined by applying (13) to \( \left\{ S^{(i)}_j, S^{(i)}_j \right\} \), forgetting any information related to the geometry of the scatterer, and forming combinations. \( \mathcal{O} \) is either \( \text{Asym} \left[ G^0 \right] \), \( \text{Sym} \left[ \mathcal{V}^{-1}_{f_i} \right] \), or \( 0 \), depending on whether the problem is absorption/material loss, scattering/radiation or extracted power from a field.

As exemplified in Sec. [III] and illustrated in Sec. [IV] the necessity of conserving reactive power imparted by the symmetric \( U_{f_i} \) constraints is crucial for accurately predicting how a particular choice of material and domain influences whether or not a family can achieve resonant response. For all cases except extracted and radiated power from an external unpolarizable source, \( \left( S^{(i)}_j \right) = \left( S^{(i)}_j \right) \). In these instances, even though (5) and (7) show that extracted and radiated power from any current source can be cast in a form similar to the corresponding initial flux problems, the inclusion of the second source image is necessary. If an unpolarizable source is taken to lie outside the domain being optimized, \( \mathcal{V}^{-1}_0 \) is defined only as a limit. Once this limit is taken, the \( G^0 \) based expressions for extracted and radiated power result, which include the introduction of the field \( \left( S^{(i)}_j \right) = \mathcal{G}_0^{\mathcal{V}^{-1}_0} \left( J \right) \) to the objective. (With \( \epsilon \) denoting the external space of the emitter and \( d \) the optimization domain.) These differences amount to the introduction of cross terms describing the interference of the fields generated by the bare and induced currents that are no longer inherently accounted for by the scattered currents at the location of the source (multiple scattering and back action). Nevertheless, the form of these problems remains like (16) up to the addition of the unalterable background contribution of \( \text{Tr} \left[ S_j \mathcal{A} \left[ G^0 \right] \right] \).

D. Solution via Duality

To solve (15) we make use of the following lemma, closely associated with the alternating direction method of multipliers [11][13] often used for solving multiply constrained convex optimization problems, commonly referred to as Lagrange duality [38].

**Lagrange Duality.** Take \( \mathcal{O} : \mathbb{R}^n \rightarrow \mathbb{R}, \mathcal{F}_j : \mathbb{R}^n \rightarrow \mathbb{R} \) and \( \mathcal{A}_j : \mathbb{R}^n \rightarrow \mathbb{R} \) to be differentiable real valued functions defining a well-posed optimization problem

\[
\begin{align*}
\max \mathcal{O} (x) \quad (x \in \mathbb{R}^n)
\end{align*}
\]

such that \((\forall k \in K) \mathcal{A}_k (x) \geq 0 \& (\forall j \in J) \mathcal{F}_j (x) = 0.\)

Take \( m^* \) to be the corresponding maximum value and \( \mathcal{O} \) to be the domain on which all constraints are satisfied. Then, for any values of \( \{ \lambda_j \} \) and \( \{ \nu_k \}, (\forall k \in K) \nu_k \geq 0 \)

\[
\begin{align*}
\max_{x \in \mathcal{O}} \left\{ \mathcal{O} (x) + \sum_{j \in J} \lambda_j \mathcal{F}_j (x) + \sum_{k \in K} \nu_k \mathcal{A}_k (x) \right\} \geq m^*.
\end{align*}
\]

Further, taking \( \mathcal{L} = \mathcal{O} (x) + \sum_{j \in J} \lambda_j \mathcal{F}_j (x) + \sum_{j \in J} \lambda_j \mathcal{A}_j (x) \) to be the Lagrangian of the optimization, the function \( g = \max_{x \in \mathcal{O}} \mathcal{L} \) is convex, and, if a set \( \{ \lambda_j \}, \{ \nu_k \} \) globally minimizing \( g \) is found such that \( \nu_k \mathcal{A}_k (x) = 0 \), where \( \tilde{x} \) is the maximum of \( \mathcal{L} \) in \( \mathcal{O} \) for \( \{ \lambda_j \}, \{ \nu_k \} \), then \( \tilde{x} \) is a solution of the original optimization problem.

**Proof.** For any point in the domain of the original (primal) optimization, \( x \in \mathcal{O} \), we have \( (\forall k \in K) \mathcal{A}_k (x) \geq 0 \), and so \( \mathcal{L} (x) \geq \mathcal{O} (x) \). Thus, \( g = \max_{x \in \mathcal{O}} \mathcal{L} \geq \max_{x \in \mathcal{O}} \mathcal{O} \) and the first statement follows immediately. Similarly, \( g \) is convex
as it is a max over affine functions of \( \{ \lambda_j \} \) and \( \{ v_k \} \). If a collection \( \{ \{ \lambda_j \}, \{ v_k \} \} \) is found such that \( \sum_{k \in K} v_k \mathcal{P}_k(\mathbf{x}) = 0 \) then \( \mathcal{A}(\{ \lambda_j \}, \{ v_k \}) = \mathcal{O}(\mathbf{x}) = \max_{\mathbf{x} \in \mathcal{O}} \mathcal{O}(\mathbf{x}) \). Thus, by the first proposition, \( \mathcal{G} \) can never be smaller than this value, and since \( \mathcal{G} \) is a convex function, \( \{ \lambda_j \} \) and \( \{ v_k \} \) globally minimize \( \mathcal{G} \). □

Whenever the operator appearing in the bilinear term is positive definite, each of the constraints in (15) (along with the objective itself) describes a compact manifold. This is always true of (12), and so, as each constraint is a dual to (13), (15) is compact. Moreover, by the validity of the \( |\mathbf{T}| = 0 \) solution, the domain is non-empty. Therefore, (15) is assured to have a unique maximum value occurring at some stationary point (or points). Due to these features, it is meaningful to consider the Lagrangian dual

\[
\mathcal{G}(\zeta, \gamma) = \max_{\mathcal{F}} \mathcal{L},
\]

where the domain \( \mathcal{F} \) is set by the criterion that max \( \mathcal{L} \) is finite. Under this assumption, taking partial derivatives over \(|\mathbf{T}|\), a stationary point of \( \mathcal{L} \) requires (\forall f_i),

\[
\left( \zeta \text{Asym}[\mathbf{U}_{f_i}] + \gamma \text{Sym}[\mathbf{U}_{f_i}] + \mathbf{O}_{f_i} \right) |\mathbf{T}_{f_i}| = \left( \frac{\zeta}{2} |\mathbf{S}_{f_i}^{(3)}| + \frac{\gamma + i\zeta}{2} |\mathbf{S}_{f_i}^{(4)}| \right)
\]

A collection \( \{ \zeta, \gamma \} \in \mathcal{F} \) if and only if \( \mathbf{O}_{f_i} + \zeta \text{Asym}[\mathbf{U}_{f_i}] + \gamma \text{Sym}[\mathbf{U}_{f_i}] \) is positive-definite for all \( f_i \), and so

\[
(\forall f_i) \mathcal{A}_{f_i} = \mathcal{A}_{f_i}^{-1} = \left( \mathcal{O}_{f_i} + \zeta \text{Asym}[\mathbf{U}_{f_i}] + \gamma \text{Sym}[\mathbf{U}_{f_i}] \right)^{-1}
\]

is both defined and positive definite. Letting \( |\mathbf{S}_{f_i}^{(3)}| = (\zeta - i\gamma) |\mathbf{S}_{f_i}^{(3)}| + |\mathbf{S}_{f_i}^{(4)}| \), and \( |\mathbf{S}_{f_i}^{(4)}| = \mathcal{A}_{f_i} |\mathbf{S}_{f_i}^{(3)}| \), it follows that \( |\mathbf{T}_{f_i}| = \frac{\zeta}{2} |\mathbf{S}_{f_i}^{(3)}| \). Hence, within \( \mathcal{F} \),

\[
\mathcal{G} = \kappa \sum f_i |\mathbf{S}_{f_i}^{(3)}|^2 + \frac{1}{4} \sum f_i \langle \mathbf{S}_{f_i}^{(3)} | \mathbf{A}_{f_i} |\mathbf{S}_{f_i}^{(3)} \rangle
\]

The gradient of (20) exactly reproduces the constraint equations.

\[
\frac{\partial \mathcal{G}}{\partial \zeta} = \mathcal{C}_\zeta = \sum f_i \frac{1}{2} \text{Re} \left[ \left( \mathbf{S}_{f_i}^{(3)} | \mathbf{S}_{f_i}^{(4)} \right) - \frac{1}{4} \langle \mathbf{S}_{f_i}^{(4)} | \text{Asym}[\mathbf{U}_{f_i}] |\mathbf{S}_{f_i}^{(3)} \rangle \right]
\]

\[
\frac{\partial \mathcal{G}}{\partial \gamma} = \mathcal{C}_\gamma = \sum f_i \frac{1}{2} \text{Im} \left[ \left( \mathbf{S}_{f_i}^{(4)} | \mathbf{S}_{f_i}^{(3)} \right) - \frac{1}{4} \langle \mathbf{S}_{f_i}^{(3)} | \text{Sym}[\mathbf{U}_{f_i}] |\mathbf{S}_{f_i}^{(4)} \rangle \right]
\]

Therefore, if a stationary point within the feasibility region is found, strong duality holds. In this case, the solution of (15) is

\[
\mathcal{O} = \sum f_i \frac{1}{2} \text{Re} \left[ \left( \mathbf{S}_{f_i}^{(3)} | \mathbf{S}_{f_i}^{(4)} \right) - \frac{1}{4} \langle \mathbf{S}_{f_i}^{(4)} | \text{Asym}[\mathbf{O}_{f_i}] |\mathbf{S}_{f_i}^{(3)} \rangle \right]
\]

with \( \{ \zeta, \gamma \} \) set by the simultaneous zero point of (21). If no such point exists in \( \mathcal{F} \), then the unique minimum value of \( \mathcal{G} \) attained on the boundary of some \( \mathcal{A}_{f_i} \), becoming semi-definite remains a bound on \( \mathcal{O} \) in (15), \( \mathcal{O} \leq \sum f_i \langle \mathbf{S}_{f_i}^{(3)} | \mathcal{A}_{f_i} |\mathbf{S}_{f_i}^{(3)} \rangle / 4 \). Comments on methods to solve (21) are given in Sec. III.

II. RELATIONS TO PRIOR ART

Previous work in the area of photonic performance asymptotics can be loosely classified into three overarching strategies: modal decompositions, exploiting quasi-normal, spectral, characteristic, Fourier and/or multipole expansions to derive per-channel performance bounds [114–128], have been broadly considered for many decades. Like the classical diffraction and blackbody limits of ray optics, channel-based bounds have proven to be of great practical value for large objects interacting with propagating waves [60, 129, 130]. However, the need to enumerate and characterize what modes may possibly participate has also long proved problematic. Small sources, separations, and domains typically require many modes to be properly represented in any basis well suited to analysis of Maxwell’s equations, and so, especially in the near-field and without knowledge of the geometric characteristics of the scattering object, there is no generally effective systematic approach to bound modal sums (without introducing additional aspects). While a variety of considerations (transparency, size, etc.) can and have been heuristically employed to introduce reasonable cut-offs [58, 67, 78, 131–133], the values obtained by modal methods in such settings are consistently orders of magnitude too large [76, 77, 134]. Still, the notion that modal descriptions often clarify otherwise unintuitive aspects of photonics remains a key insight.

Shape-independent conservation limits, utilizing energy [59, 75, 77] and/or spectral sum rules [83, 104, 135, 140] to set local limits based on global physical laws, generally display the opposite behavior, and are known to give highly accurate estimates of maximal absorption and far-field radiative emission phenomena in the limit of vanishingly small size (quasi-static) for certain metallic objects [9, 10, 77, 139, 141]. Notwithstanding, as we have found in our work on bounds for radiative heat transfer [73, 78] and angle-integrated radiative emission [74], they are not sufficiently in and of themselves to properly capture various relevant and performance-limiting wave effects. Intuitively, without any geometric information, a conservation argument must apply on a per-volume basis, which is at odds
with the area scaling of ray optics.

As a relevant example, consider the global requirement that the power quantities given in section 1 must be non-negative. Two of these turn out to be unique and thus set physically motivated algebraic constraints on the T operator. For any vector field $|E|$, the positivity of scattering (known as passivity [142]) imposes

$$\langle E| \text{Asym}[T] - T^\dagger \text{Asym}[V^{-1}] T|E\rangle \geq 0,$$

while the positivity of absorption imposes

$$\langle E| \text{Asym}[T] - T^\dagger \text{Asym}[G^0] T|E\rangle \geq 0.$$

Both of these conditions are included and strengthened in [12], which amounts to a statement of the optical theorem [109]. The sum of the absorbed power and scattered power, and $|\psi|$, must be equal to the extracted power, and. If no additional restrictions pertaining to possible geometry or the characteristics of $|E|$ are given, then the most that can be said is that no singular value of $|T|$ can be larger than the inverse of the smallest singular value of $\text{Asym}[V^{-1}]$. For a local electrical medium of permittivity $\chi$, this logic implies a bound on polarization response $|\langle T\rangle| \leq \zeta_\text{mat}$ described by the material-loss figure of merit,

$$\zeta_\text{mat} = |\chi|^2/\text{Im} [\chi]$$

originally derived in Ref. 127 using the implications of passivity for polarization fields. The universal applicability of this largest possible response has profound consequences for the design of many photonic devices relying on weakly metallic response ($\approx -10 < \text{Re}[\chi] < -2$) and small interaction volumes [67, 141]. In these cases, it is often fair to assume that a resonance can be created and that $\text{Asym}[G^0] \approx 0$, as the maximum achievable polarization current is indeed dominated by material losses. But, for devices where light-matter interactions occur on length scales comparable to or greater than the wavelength $(\gtrsim \lambda/10)$, or the real part of $\chi$ is outside the range given above, such estimates are overly optimistic for single material devices. Over a large enough domain, the generation of polarization currents capable of interacting with propagating fields necessitates radiative losses which have been neglected by supposing $\text{Asym}[G^0] \approx 0$. In order to create an active far-field resonance, it must be possible to couple to radiation modes and then confine the resulting generated field within the domain.

Scattering operator approaches broadly aim to eliminate the weaknesses of modal and shape-independent conservation arguments by combining their strengths [73, 74, 78, 105, 143, 149]. Innately, the Green's function of an encompassing domain (through its link to Maxwell's equations) provides both a modal basis for, and constraints on, modal sums. Restrictions on the possible characteristics of the T operator can be used to ensure that physical laws and scaling behavior are observed. A number of encouraging conclusions have been derived in this manner. Drawing from our own work, in Ref. 73 it was shown that imposing $|\langle T\rangle| \leq \zeta_\text{mat}$ on the operator expression for angle-integrated absorption and thermal emission, is sufficient to generate bounds smoothly transitioning from the absorption cross-section limits applicable to resonant metallic nanoparticles (the aforementioned shape-independent conservation arguments) to the macroscopic blackbody limits of ray optics. Similar methods were used in Ref. 78 to prove that, for equal values of $\zeta_\text{mat}$, nanostructuring cannot appreciably improve near-field thermal radiative heat transfer compared to a (simple) resonant planar system. Nevertheless, careful investigation of the previous situations where scattering operator amalgamations have been successfully applied shows a consistent use of niceness properties that are not always present. In the examples given above, we were aided by the fact that thermal sources are completely uncorrelated and, for thermal emission and integrated absorption, that only propagating fields needed to be treated. Without these helpful facts there are situations where past scattering operator approaches, which from focused exclusively on focused exclusively on [12], would add complexity without tightening the asymptotics provided by shape-independent conservation arguments (dashed lines of the figures in Sec. IV). Moreover, using the standard technique of translating established physical principles back to implied operator properties and then using inequality compositions to produce limits, it is difficult to see how the interaction of more than one or two additional constraints could be properly accounted for. The flexibility offered by Lagrange duality, suggests that this view may be of substantial benefit going forward.

A related shift towards systemization has been realized in the recent report of computational bounds by Angeris, Vučković and Boyd [69], providing a single framework for any linear optimization problem in terms of a target field (or a collection of target fields). The result, also making use Lagrange duality, has immediate consequences for heuristically understanding and improving inverse design. Yet, it does not allow one to make conclusive statements about feasibility and relative performance as is true of traditional limits. More properly, what is found is a “computational certificate”: given a target field and an evaluation metric, the algorithm returns a number; any vector satisfying Maxwell's equation will have a metric disagreement with the target at least as large as the number. That is, the algorithm does not find physical limits, but instead a minimum bound on distance, in a certain user determined measure, between a particular field and the set of physically possible fields. There may be certain situations where this difference is of little consequence, or provably zero, but a priori there are no guarantees. There need not be any relation between the value taken by an objective at a point and how near that point is to some set.

Finally, while concluding the writeup of this article, we have become aware of the recent work of Gustafsson et al. [69], extending contemporary methods for determining bounds on macroscopic antenna performance to photonics [9, 19, 69, 70]. Independently developed, the formulation is in many respects equivalent to the method we have
presented. Working from the perspective of polarization currents, the optimization of objectives equivalent to (4–8), subject to global power constraints equivalent to (12) and (13), is undertaken via minimization of the Lagrangian dual. This leads to a rich collection of findings reinforcing some of the insights found in Sec. IV.

III. COMPUTATIONAL MECHANICS AND SINGLE CHANNEL ASYMPTOTICS

To present the conceptual mechanics of (15), in this section we detail our computational procedure. The discussion is broken into two subsections. The first outlines the general method by which all results below are obtained. The second considers a simplified single-channel problem that becomes exact in situations comprising small domains or dominated by high angular momenta. These solutions herald features observed in Sec. IV.

A. Computational Mechanics

We begin with some obligatory introduction of notation and clarification of how the \( \mathbf{U}_f \) and \( \mathbf{A}_f \) matrices central to (15) and (15) can be obtained. These atomic elements are found to be a considerable help to later analysis.

Recall that the Green’s function can always be expanded in terms of the “regular” (finite at the origin), \( \mathbf{RN} \) and \( \mathbf{RM} \), and outgoing, \( \mathbf{N} \) and \( \mathbf{M} \), spherical wave solutions to Maxwell’s equations as \[ G^0(x, y) = -\int \delta(x - y) \hat{x} \otimes \hat{y} + \sum_{m} (-1)^m \left[ \mathbf{M}_{f,m}(x) \mathbf{RM}_{f,m}(y) + \mathbf{N}_{f,m}(x) \mathbf{RN}_{f,m}(y) \right], \quad x > y \]

\[ \left[ \mathbf{M}_{f,m}(x) \mathbf{RM}_{f,m}(y) + \mathbf{N}_{f,m}(x) \mathbf{RN}_{f,m}(y) \right], \quad x < y. \tag{24} \]

Here, \( x \) and \( y \) are used to denote the wavevector normalized radial vectors of the domain and codomain, i.e. \( x = \langle 2\pi r / \lambda, \theta, \phi \rangle \), with \( x \) and \( y \) denoting their corresponding radial parts, and the integral over \( y \) is taken to mean integration over the \( y \) coordinate. (Note that there are no complex conjugation in these integrals.) The sum over the magnetic number \( m \) runs from \(-\ell\) to \( \ell \), and the sum over the angular momentum number \( \ell \) begins at 1. (Our notation for the Green function is unconventional in that an additional factor of \( k_0^2 = (2\pi^2 / \lambda^2 \) is included as part of the definition.)

So long as the current source is not located within the domain in question, any resulting incident field can be expanded in terms of the regular waves [106] [150] [151]. Hence, the spectral basis of the asymmetric part of (24)

\[ \text{Asym}[G^0] = \sum_{\ell, m} (-1)^m \int \mathbf{M}_{f,m}(x) \mathbf{RM}_{f,m}(y) + \mathbf{RN}_{f,m}(x) \mathbf{RN}_{f,m}(y), \tag{25} \]

the unit normalized \( \mathbf{RN}_{f,m} \) and \( \mathbf{RM}_{f,m} \), serves as convenient choice for generating the \( f \) basis vector families appearing throughout the article. That is, given the form of the regular solutions

\[ \mathbf{RN}_{f,m}(y) = \frac{\sqrt{\ell + 1}}{y} j_\ell(y) A^{(2)}_{f,m} + \frac{1}{y} \frac{\partial}{\partial y} [y j_\ell(y)] A^{(2)}_{f,m}, \]

\[ \mathbf{RN}_{f,m}(y) = j_\ell(y) A^{(2)}_{f,m}, \quad \mathbf{RM}_{f,m}(y) = y j_\ell(y) A^{(1)}_{f,m}, \] \tag{26}

the orthonormality of the vector spherical harmonics ( \( A^{(1)}_{f,m}, A^{(2)}_{f,m} \), and \( A^{(3)}_{f,m} \), see Ref. 153 for details) means that the Green function (24) does not couple the \( f \), \( m \) or \( \mathbf{RN} \) and \( \mathbf{RM} \) labels, and so the individual radiation channels act as an effective partitioning. By then taking these vectors as the family heads, a complete (simplifying) basis for (15) can be generated through the Arnoldi (Krylov subspace) procedure [154].

Briefly, starting with a given unit normalized regular wave, \( \mathbf{RN}_{f,m} \), one generates \( \mathbf{U} \mathbf{RN}_{f,m} = (\mathbf{V}^{-1} - \mathbf{G}^{0}) \mathbf{RN}_{f,m} \). Projecting out the \( \mathbf{RN}_{f,m} \) component of this image and normalizing, one obtains a new vector \( \mathbf{PN}^1_{f,m} : \mathbf{PN}^{(2)}_{f,m} \) then serves as the input for the next iteration, and in this way the \( f \) block, more properly the \( \mathbf{RN}_{f,m} \) block, of the matrix representation of the \( \mathbf{U} \) operator (\( \mathbf{U}_f \)) is computed. The \( k \) labels appearing elsewhere in the article are defined to run over the this Arnoldi basis, \( \{ \mathbf{RN}_{f,m}, \mathbf{PN}^{(2)}_{f,m}, \mathbf{PN}^{(3)}_{f,m}, \ldots \} \), and each \( \mathbf{U}_f \) results from the associated representation of \( \mathbf{U} \). Technically the above process does not terminate, but regardless, three practical consideration lead to numerical convergence [155].

First, due to the fact that each vector is orthogonal to all others, the off diagonal coupling components of \( \mathbf{U}_f \) originate entirely due to the volume integrals in (24). Therefore, as seen in the next subsection, in the limit of vanishing volume or high \( \ell \), each \( \mathbf{U}_f \) is effectively \( 2 \times 2 \). Second, by the Arnoldi construction, all upper diagonals beyond \( \text{diag}_1 \), with \( \text{diag}_0 \) standing for the main, are zero. Hence, because \( \mathbf{U} = \mathbf{U}^* \) (as the operators entering its definition are reciprocal), its matrix representation in our chosen basis is tridiagonal. Third, because any source can be represented solely in terms of the regular waves [106], full numerical convergence is not actually required. \( \mathbf{U}_f \) blocks enter (15) in two ways: as inverses through the \( \mathbf{A}_f \) blocks, and directly through the constraints (21). This gives a simple criteria on the size of the basis needed to achieve numerical convergence. Once the solution of the banded linear system of equations \( \mathbf{A}_f^{-1} \) achieves convergence for a source vector that is zero in all entries but the first [155], the Arnoldi basis of \( f \) is sufficiently large. This procedure need only be done once for each \( f \) in a given problem by enforcing superficially strong termination criteria on an initial test problem. The banded nature of \( \mathbf{A}_f^{-1} \) also provides a simple, conclusive, estimate of the error. Simply, all that is required is to pad the current solution and calculate its image under \( \mathbf{A}_f^{-1} \) in a basis augmented by three additional elements. The magnitude of the error of the image compared to the source is exactly the same as would be found.
in any larger (even infinite) basis, see Sec. VII for additional details.

Under this umbrella, the validity of (21) maps the determination of bounds for any electromagnetic interaction that can be described as a total absorption, scattering, or extinction process, to the determination of the minima of a constrained convex function, (20). Many efficient algorithms exist to solve such problems [157][159], along with a variety of excellent introductions [38][154][160].

B. Single Channel Asymptotics

To give a better description of (15), we now carry out a single channel (family) optimization on a spherically bounded domain of radius \( R \) (here taken to be the product of the true radius and the wavevector). This simplified problem captures the primary features observed in Sec. IV in the limits of vanishing \( R \) (quasi-static) or dipole separation \( d \). In either case, the largest possible interaction of \( T \) with the propagating field, over the containing ball, is found to obey a relation very close to the material dependence encountered in Rayleigh scattering [73]. For low loss dielectrics (Re\( \chi > 0 \)) and strong metals (Re\( \chi < -3 \)), this can lead to large discrepancies with respect to the previously established per-volume bounds based on the material loss figure of merit \( \zeta_{\text{mat}} = \frac{\chi^2}{\text{Im} \chi} \) [27].

Consider the optimization problem

\[
\max \left\{ \left| T^{(1)}_{\ell} \right| \right\} \text{ such that }
\begin{align*}
\zeta^*_\ell &= \text{Im} \left[ \left( S^{(1)}_\ell \right) \left( T^{(1)}_{\ell} \right) \right] - \left\{ T^{(1)}_{\ell} \right\} \text{Asym} \left[ U_{\ell} \right] \left| T^{(1)}_{\ell} \right| = 0, \\
\zeta_{\ell} &= \text{Re} \left[ \left( S^{(1)}_\ell \right) \left( T^{(1)}_{\ell} \right) \right] - \left\{ T^{(1)}_{\ell} \right\} \text{Sym} \left[ U_{\ell} \right] \left| T^{(1)}_{\ell} \right| = 0,
\end{align*}
\]

(27)

where it has been assumed that that \( \forall f_i \neq f_j \) \( \left\{ S_{f_i} \right\} = 0 \), so that (15) reduces to a problem involving a single radiative channel. Like prior \( P \) operators, here \( P_1 \) represents the projection of \( T \) onto the \( f_1 \) family head. So stated, (27) represents the maximum possible interaction that can occur between an object and a single radiative mode, as allowed by (12) and (13). Based on the power series representation of the spherical Bessel functions,

\[
j_y(r) = \sum_{q=0}^{\infty} \frac{(-1)^q}{q! (2l + 2q + 1)!!} \left( \frac{r}{2} \right)^{2q},
\]

there are two situations in which this problem is amenable to analytic manipulations. If either the radius \( R \) is small, compared to \( 2\pi/\lambda \), or \( \ell \) is large, compared to \( R \), then both the regular and outgoing waves appearing in the Green’s function, (24), are well approximated by two-term expansions, and the Arnoldi procedure for basis generation described above terminates after constructing a single image vector. Symbolically carrying out the required steps, one finds that the representation of \( U_{f_1} \) in the quasi-static (\( R \to 0 \)) regime is

\[
U_{f_1} = \mathcal{V}^{\ell - 1} - C^{\ell \ell^*}_{f_1} = \text{Sym} \left[ U_{f_1} \right] + i \text{Asym} \left[ U_{f_1} \right] = \\
\begin{bmatrix}
\frac{1}{3} + \text{Re} \left[ \frac{\xi_{\ell}}{\xi_{\ell}} \right] - \frac{3}{5} & \frac{2}{5} & - \frac{2}{5} \\
\frac{2}{5} & \frac{3}{5} & \frac{1}{5} \\
\frac{1}{5} & \frac{1}{5} & \frac{1}{5}
\end{bmatrix}
\begin{bmatrix}
\frac{\text{Im} \xi_{\ell}}{\xi_{\ell}} + \frac{3}{5} \\
\frac{3}{5} \\
\frac{3}{5}
\end{bmatrix},
\]

(28)

while for large \( \ell \) (high angular momentum), the representation of \( U_{f_1} \) is

\[
U_{f_1} = \mathcal{V}^{\ell - 1} - C^{\ell \ell^*}_{f_1} = \text{Sym} \left[ U_{f_1} \right] + i \text{Asym} \left[ U_{f_1} \right] = \\
\begin{bmatrix}
\frac{1}{3} + \text{Re} \left[ \frac{\xi_{\ell}}{\xi_{\ell}} \right] - \frac{3}{5} & \frac{2}{5} & - \frac{2}{5} \\
\frac{2}{5} & \frac{3}{5} & \frac{1}{5} \\
\frac{1}{5} & \frac{1}{5} & \frac{1}{5}
\end{bmatrix}
\begin{bmatrix}
\frac{\text{Im} \xi_{\ell}}{\xi_{\ell}} + \frac{3}{5} \frac{2\ell + 1}{5(\ell + 3/2)} \\
\frac{3}{5} \\
\frac{3}{5}
\end{bmatrix},
\]

(29)

where \( a \) is a constant on the order of 0. Within their respective regimes of validity, the two matrices share many features. Due to the identity portion of the Green’s function, the (1,1) elements have a constant positive piece in addition to the \( \text{Re} \left[ \xi_{\ell} \right] / \xi_{\ell} \) contribution made by \( \mathcal{V}^{\ell - 1} \). Hence, the (purely real) material response factors needed to reach resonant response are \( \text{Re} \left[ \xi_{\ell} \right] = -3 \) for quasi-statics (the localized plasmon-polariton condition for a spherical nanoparticle) and \( \text{Re} \left[ \xi_{\ell} \right] = -2 \) for high angular momentum (the surface plasmon-polariton condition for a half space [80]). Denoting the symmetric and anti symmetric components of either representation as \( u^{(1)}_{\ell} \) and \( u^{(2)}_{\ell} \), (15) translates to determining the \( \{ t_1, t_2 \} \) component pair producing the largest magnitude \( t_1 \) such that

\[
\sin(\theta) t_1 t_1 - t_1^2 u^{(1,1)} - t_1^2 u^{(2,2)} = 0
\]

\[
\cos(\theta) t_1 t_1 - t_1^2 u^{(1,1)} - t_1^2 u^{(2,2)} = 2\cos(\phi) t_1 t_2 u^{(1,2)} = 0.
\]

(30)

In (30), the \( t_1 \) and \( t_2 \) variables are the (positive) magnitude coefficients of \( |T| \) in the first and second Arnoldi family vectors; \( s_1 \) is the coefficient of the source; \( \theta \) is the relative phase difference between the source and first coefficient of \( |T| \), and \( \phi \) is the relative phase difference within the two coefficients of \( |T| \). As a response operator, \( \text{Asym} \left[ T \right] \) must be positive semi-definite and so \( \theta \in [0, \pi] \). Using the symmetric constraint to solve for \( t_2 \) in terms of \( t_1 \), forgetting \( u^{(2)} \) terms when they appear as sums against larger components in the resulting quadratic equation, the asymmetric constraint determines

\[
t_1 = \frac{\cos(\theta) t_1 u^{(2,2)} - \sin(\theta) u^{(2,2)}}{u^{(1,2)} u^{(1,1)} - u^{(1,1)} u^{(2,2)}},
\]

(31)

subject to the condition, resulting from the requirement that \( t_1 \) and \( t_2 \) are real, that

\[
(\sin(\theta) u^{(1,1)} - \cos(\theta) u^{(1,1)})(\cos(\theta) u^{(2,2)} - \sin(\theta) u^{(2,2)}) \geq 0.
\]

(32)
\[
\frac{t_1}{s_1} = \zeta_{\text{eff}} \leq \begin{cases}
\frac{1}{\delta_c} \frac{|x|}{\Im [x]} & \text{Re} [x] < 0 \ & \text{&} \ |\text{Re}[x]/|x|| \leq \delta_G, \\
0 & \text{else}
\end{cases}
\]

(33)

where the geometry and family dependent \(\delta_G\) has been introduced as the constant part of \(u_{s1}^{(1,1)} - u_{s1}^{(2,2)}\). This name is chosen as \(\delta_G\) originates from the delta function portion of (23), with

\[
\delta_G = \begin{cases}
1/3, & R \to 0 \quad \text{(quasi-statics)} \\
1/2, & \ell \to \infty \quad \text{(high momentum)}
\end{cases}
\]

Intuitively, (33) captures the fact that there is a trade-off in achieving resonance from nanostructuring a body. As consistency with our prior assumptions requires that the domain is well within the validity of the quasi-static approximation, any material structuring will, at best, alter the effective medium parameters of the domain [82–84, 163]. The first form of (33) can be understood as dilution of the material through the addition of vacuum regions until the effective response reaches the resonance condition (\(\text{Re} [x_{\text{eff}}] = -3\) for spherical nanoparticles) [81, 164]. Plugging this dilution factor into the per-volume material-loss factor \(\zeta_{\text{mat}}\) yields the effective material response,

\[
\zeta_{\text{eff}} = \text{Re} \left[ \frac{z_{\text{res}}}{X} \right] \frac{|X|^2}{\Im [X]},
\]

(34)

corresponding to the first term of (33). This homogenization material figure of merit is commonly encountered in discussing the potential of different material options for plasmonic applications [165–166].

Equation (33) already yields substantially tighter asymptotics than those predicted by the material-loss figure of merit \(\zeta_{\text{mat}} = |X|^2 / \Im [X]\) for low loss dielectrics (\(\text{Re} [X] > 0, \text{Im} [X] \ll 1\)) or strong metals (\(\text{Re} [X] \ll -\delta_G\)). The minimum of (33) is found to accurately predict the characteristics of the planewave bounds of (15) as \(R \to 0\), Fig. 1. Quasi-static absorption, (4), is well described by \(4\pi\zeta_{\text{eff}} R^3/3\); while scattering, by (5), \(\langle T | \text{Asym} [G^0] | T \rangle\), is approximately \(8\pi\zeta_{\text{eff}} R^6/27\). While the monotonicity property of the framework means that these results (properly scaled) will hold for any domain geometry, and one may reasonably guess that the characteristics of the Arnoldi process on which the above arguments rest are similar in any small volume limit, it should be kept in mind that other domain geometries (e.g., ellipsoids [27, 139]) may well display stronger response per unit volume. That is, while the polarization field may have a much larger interaction with an incident plane wave within the volume of a nanostructured object, the net enhancement will be weaker than the minimum of the \(\zeta_{\text{eff}}\) given above once the ratio of its volume to an encompassing ball is accounted for.

IV. APPLICATIONS

In this section, we discuss applications of (15) to two canonical situations of practical and conceptual value: limits on absorbed and scattered power for a planewave incident on any structure contained in a ball of radius \(R\), and limits on the radiative power that can be extracted from a dipolar current source separated from the same domain by a distance \(d\). The former examples characterize the familiar scattering cross sections of bodies [108] while the latter are crucial to the characterization of local field enhancements [27, 167] and light–matter interactions [168, 169] in structured media. (By Poynting’s theorem, the radiated power enhancement figure of merit shown in Fig. 4 is the radiative part of the well-known Purcell enhancement factor [50].) By symmetry, evaluation of the bounds for a spherical boundary is simplest (analytically) in the basis of spherical harmonics \(A^l_j\). Throughout the section \(R\) is unnormalized unless otherwise stated, and \(\ell\) will stand for the angular momentum number.

A. Plane Wave Source

For an incident planewave, the magnitude of each field coefficient is strongly tethered to the radius of the domain through the relation

\[
\hat{E}_i = \sum_{l=0}^{\infty} \sum_{m=1}^{21} i^{l+1} \sqrt{(2l+1)\pi} \text{RM}_{l,1,1}(r, \theta, \phi) \pm i^{l+1} \sqrt{(2l+1)\pi} \text{RN}_{l,1,1}(r, \theta, \phi).
\]

(35)

where \(\text{RM}_{l,\pm 1}(r, \theta, \phi)\) and \(\text{RN}_{l,\pm 1}(r, \theta, \phi)\) are the regular (finite at the origin) solutions of Maxwell’s equations as given in (26), and \(r\) is the wavevector normalized radial component (the product of the true and \(2\pi/\lambda\)). This leads to a complementary action of the global power conservation constraints. For any particular combination of material and radius, other than a resonant quasi-static object, even without the further suppression caused by the per-channel constraints it is not possible to achieve the ultimate material limit discussed in Sec. III. \(\|T\| \leq \zeta_{\text{mat}} = |X|^2 / \Im [X]|.\) In the quasi-static limit, the behavior of the bounds is completely captured by the analysis of Sec. III. Throughout, all solutions, excluding outlier points of numerical instability, are found to be strongly dual. Note that for small radii, the dashed curves considering only the global conservation of real power exactly match what would be seen at the quasi-static resonance condition of \(\text{Re} [X] = -3\).

Quasi-Statics—As detailed in Sec. III B, the requirement that both real and reactive power be simultaneously conserved, ultimately on a per-channel basis, has far-reaching implications for scattering interactions in small domains. In particular, (13) acts to incorporate phase information on top of the maximum polarization magnitude constraint set by (12), such that when both constraints are taken into account it is no longer true that the per-volume material-
FIG. 2. Bounds on absorption cross-sections for incident planewaves. The panels show bounds on the maximum power that can be absorbed from an incident planewave of wavelength $\lambda$ by an arbitrarily shaped object contained in a ball of radius $R/\lambda$ (normalized by its geometric cross section), for representative values of the object’s electric susceptibility, $\chi$. The dashed lines result from solving (15) for (4) using only conservation of power (12) as a constraint. The dotted lines result from imposing conservation of reactive power (13) as an additional constraint. Small radii features are in exact agreement with the asymptotic predictions of Sec. III, revealing a diluted (homogenized) quasi-static enhancement for materials that deviate from the resonant (localized plasmon-polariton) condition associated with a spherical nanoparticle, $\text{Re}\chi = -3$. As $R \to \infty$, the geometric cross-section limit of ray optics is recovered. For small $R$, the dashed curves corresponding to $\text{Re}\chi = 3$ in panel (a) precisely reproduce the bounds for $\text{Re}\chi = -3$, consistent with the per-volume material scaling $\propto |\chi|^2/\text{Im}\chi$ expected from only enforcing (12).

loss figure of merit $\zeta_{\text{mat}}$ holds. More precisely, $\zeta_{\text{mat}}$ scaling occurs uniquely, for a spherically bounded domain under the resonant condition $\text{Re}\chi = -3$ corresponding to the plasmon-polariton condition of a spherical nanoparticle [80]. For all other materials, markedly different characteristics arising from (33) are observed. For dielectrics, $\text{Re}\chi \gg -3$, the resonant material scaling is inverted $\propto (\zeta_{\text{mat}})^{-1}$, so as to produce material scaling that can in fact increase with larger loss (consistent with non-resonant asymptotics). For strong metals, $\text{Re}\chi \ll -3$, an effective (homogenized) scaling of (34), with $\text{Re}\chi_{\text{res}}$ the (geometry dependent) value of $\text{Re}\chi$ at which the first radiation mode enters resonance. Such scaling captures the difficulty for achieving resonance and coupling to far field radiation fields, and the tradeoff with respect to the requisite dilution of the medium (through the addition of vacuum regions) until the effective response is resonant (with $\text{Re}\chi_{\text{res}} = -3$ for spherical domains).

Wavelength Scale—At radii approaching the wavelength scale ($R \gtrsim \lambda/10$), the strict validity of the quasi-static argument given in Sec. III B becomes increasingly tenuous. The growth of source weight into new scattering channels opens the possibility of utilizing a wider range of wave physics (e.g. leaky and guided resonances [170,171]), while the concurrent weakening of reactive power conservation requirements and strengthening of real power conservation requirements leads to a more intricate interplay of the constraints in (15). Together, these factors are responsible for the distinct features observed in the dielectric examples of Figs. 2 and 3: as resonance conditions (effectively $R \approx$...
FIG. 3. **Bounds on scattering cross-sections for incident planewaves.** The panels show bounds on the maximum power that can be scattered from an incident planewave of wavelength $\lambda$ by an arbitrarily shaped object contained in a ball of radius $R/\lambda$ (normalized by its geometric cross-section) and of electronic susceptibility $\chi$. The dashed lines result from solving (15) for (5) using conservation of real power (12) as a constraint. The dotted lines result from further imposing conservation of reactive power (13) as an additional constraint. As $R \to 0$, all plots agree with the quasi-static predictions of (33). Markedly larger enhancements $\propto |\chi|^2/\text{Im}[\chi]$ may be achieved for materials satisfying the resonant (localized plasmon-polariton) condition of a spherical nanoparticle, $\text{Re}[\chi] = -3/2$. 

$1/(2\sqrt{\text{Re}[\chi]})$ are satisfied for higher angular momentum channels modal bumps appear. In particular, as the spherical boundary expands, channel by channel, the smallest eigenvalue of $\text{Sym}[U_\ell]$ continually shrinks until the operator eventually loses definiteness, causing (13) to loosen. This behavior is first observed in $\ell = 1$, with the initial increase and subsequent hold originating from the largest singular value of $\text{Sym}[U_\ell]$ decreasing rapidly before stabilizing around the half wavelength condition of the spherical Bessel function $(\text{min } r) \equiv \partial j_1(2\pi r/(\lambda \sqrt{\text{Re}[\chi]}))/\partial r = 0$. The second peak occurs near the full wavelength condition, $(\text{min } r) \equiv j_1(2\pi r/(\lambda \sqrt{\text{Re}[\chi]})) = 0$, which coincides with a loss of definiteness in $\text{Sym}[U_\ell]$. Higher peaks follow a similar pattern. First, there is the an initial spike corresponding to the half wavelength peak. This is then succeeded by a sharp edge occurring near the full wavelength condition. However, at the same time, the inflation of the boundary also leads to an increase in necessary radiative losses (as discussed in Sec.[7] and Ref.[74]), which is manifested in the operators as an increase in the value of $\langle T_\ell |\text{Asym}[U_\ell] | T_\ell \rangle$. The presence of these additional contributions restricts the magnitude of $\langle T_\ell |\text{Asym}[U_\ell] | T_\ell \rangle$ to a geometry dependent value below the figure of merit $\zeta_{\text{mat}}$, and so rather than completely releasing to this larger enhancement value, the bound slips and catches.

**Ray Optics**—Past $R \gtrsim \lambda$, the achievable scattering interaction in any given channel is increasingly dominated by the value of $\langle T_\ell |\text{Asym}[U] | T_\ell \rangle$. As such, the bounds calculated by asserting (12) as the only power constraint on an otherwise free optimization problem (Fig. 2 and Fig. 3 dashed lines) are observed to be sufficient. Making this reduction, (15) becomes congruous to the thermal radiation problem we have considered in Ref.[74]. The planewave expansion coefficients of (35) exhibit exactly the same per-channel characteristics considered in that article, and so,
FIG. 4. Bounds on radiative emission from dipole sources. The panels show bounds on the radiative power (normalized by vacuum radiative emission) that can be extracted from a dipolar current source oscillating with frequency $2\pi c / \lambda$ and polarized perpendicular to a spherical bounding domain of radius $R = \lambda / 2$, as a function of their mutual separation $d$ and for representative values of the object's electric susceptibility, $\chi$. The dashed lines are determined by imposing only the conservation of real power (12). The dotted lines result from further imposing conservation of reactive power (13) as an additional constraint. A scaling between diluted (homogenized) effective response, described by (34), and the material figure of merit $\zeta_{\text{mat}}$ in (23) is observed, leading to bounds that, depending on $\chi$, can differ by orders of magnitude. Asymptotic scaling $\propto d^{-3}$ is observed in the deep near-field regime of $d / \lambda \rightarrow 0$.

because (12) is asymptotically equal to the absorption objective, the same asymptotic behavior is encountered. Regardless of material parameters, each of the scattering objectives (3)–(8) begins to scale as the geometric cross section of the bounding sphere once the radius is taken to be sufficiently large. For absorption, this leads to a value equal to the geometric cross section of the ball, $\pi R^2$. For extinction and scattering, a value of $4\pi R^2$ is found, two times larger than what would be expected based on the extinction paradox [172] [173]. The genesis of this additional factor is presently unknown.

B. Dipole Source

The field of dipole oriented perpendicular to spherical domain of radius $R$, separated by distance $d$, is [151]

$$\mathbf{E}_\perp (r, R, d) = \frac{i k}{\sqrt{8\pi}} \sum_{\ell} \sum_{\nu = \ell - 1}^{\ell + 1} (2\nu + 1) \frac{2 + \ell' (\ell' + 1) - \nu (\nu + 1)}{2} ^{1 - \nu - \ell'} \left( \begin{array}{ccc} 6\ell + 3 & 0 & 0 \\ 0 \ell' & 0 \end{array} \right) \mathbf{h}_\nu^{(1)} (R + d) \mathbf{R} N_{\ell,0} (r),$$

(36)

where $\mathbf{h}_\nu^{(1)} (R + d)$ is the first spherical Hankel function, $d$ is the wavevector normalized distance of the dipole from the spherical boundary and $R$ is the wavevector normalized radius of the bounding sphere. The limits of the sums in this expression are set explicitly based on the Wigner-3j selection rules. Originating from this more complicated coefficient dependence, the interaction features of the dipole
are found to mix various aspects of the three length scales discussed for an incident planewave. Figure 4 explores scattered power from a dipole in the vicinity of a ball of radius \( R = \lambda/2 \) (wavelength scale). Noticeably, the \( \propto d^{-3} \) growth of the field within the ball results in a corresponding asymptotic scaling at small separations. However, revisiting the planewave results of Fig. 2 and Fig. 3 suggests that for a radius \( R = \lambda/2 \), a susceptibility value of \( \Re[\chi] = 16 \) allows a greater number of low \( \ell \) modes to achieve resonance compared to \( \Re[\chi] = 4 \), and this has an impact on the observed material dependence. In particular, the scaling of the bounds with respect to \( \chi \) shares some qualitative similarities with the observed behavior of Sec. III in quasi-static and high \( \ell \) settings. In particular, an approximately diluted (homogenized) scaling with respect to the material figure of merit \( \zeta_{\text{mat}} \) is observed in the deep near-field regime \( d/R \ll 1 \), owing to the aforementioned penalty in achieving resonant behavior within the domain through nanostructuring. The precise nature of the dilution in near-field situations, however, remains an open question requiring analysis beyond the single-channel asymptotics of Sec. III. Effectively, because extraction of radiative power from a dipole in the near field of the domain requires coupling between “radiative” and “non radiative” channels, the relative strength of near field (large \( \ell \)) enhancement is not the only relevant factor driving overall performance. From (29), in the \( (\ell \to \infty) \) limit the asymmetric part of \( U_f \) associated with coupling to radiation \([74]\) is \( \propto \frac{\pi(\ell+1)/\pi R/2^{\ell+1}}{[\ell+3/2]^2} \), which tends to zero rapidly as \( \ell \) increases. Therefore, assuming that the radiative problem can be purely described by large-\( \ell \) characteristics invariably leads to the erroneous conclusion that no radiative enhancement is possible.

V. SUMMARY REMARKS

The ability of metals and polaritonic materials to confine light in subwavelength volumes without the need for any other surrounding structure (plasmon–polaritons \([174, 175]\), coupled with the variety of geometric wave effects achievable in dielectric media (band gaps \([176, 177]\), index guiding \([178, 179]\), topological states \([180, 181]\)), rest as bedrocks of contemporary photonic devices. Yet, the relative merits and potential of these two overarching design approaches for controlling light–matter interactions remains a widely studied \([182, 184]\) topic. The broad strokes are well established. The subwavelength confinement and large field enhancements offered by metals are offset by the fact they are fundamentally tethered to substantial material loss \([183]\). Through interference, dielectrics architectures may also confine and intensify electromagnetic fields, and can do so without large accompanying material absorption \([51]\). But, accessing these effects invariably requires larger domains. Within rigidly specified designs, comparisons between different such subclasses have been made \([64]\). Yet, it remains an open question whether additional nanostructuring or material freedom may lead to entirely different tradeoffs and/or conclusions. As with the rising questions of limits for computational methods highlighted in the introduction, a central driver of debate is a lack of concrete (pertinent) knowledge of what is possible, beyond qualitative arguments.

We believe that the simple canonical examples shown in Sec. IV provide compelling evidence that the Lagrange duality framework we have described in this article offers a path towards progress. As verified in Sec. IV of this work, the consistency of the scheme is amenable to numerical evaluation under realistic photonic settings (for practical domain sizes and materials) and sufficiently broad to provide both quantitative guidance and physical insights: as the size of an object interacting with a planewave grows, there is a transition from the volumetric scaling characteristic of subwavelength objects to the geometric cross-sectional dependence characteristic of ray optics; critical sizes exist below which it is impossible to create dielectric resonances; material loss dictates achievable interactions strengths only once it becomes feasible to achieve resonant response and in turn, significant coupling to the source. Surprising insights were also obtained: even under complete freedom over structuring, strongly metallic \( \Re[\chi] \ll 0 \) subwavelength objects \( (R \ll \lambda) \) are unable to match the scattering response of spherical nanoparticles satisfying the localized plasmon-polariton resonance condition, \( \Re[\chi] = -3 \), for comparable values of material loss, \( \Im[\chi] \), instead exhibiting a diluted (homogeneized) response \( \propto \Re[\chi]/\Im[\chi] \). Effectively, the conservation of reactive power and associated constraints on achievable phase (resonant response) strongly limits the amount of power that can be extracted from either a planewave or a dipolar source above a spherically bounded object.

Translating the above framework beyond the spectral bases employed here onto a completely geometry-agnostic numerical algorithm will make it possible to analyze the realative tradeoffs associated with various kinds of optical devices. By varying material and domain parameters, the significance of different design elements from the perspective of limit performance can be ascertained in a number of technologically relevant areas. The basic scattering interaction quantities we have considered, Sec. I, rest at the core of engineering the radiative efficacy of quantum emitters \([66, 68]\), resonant response of cavities \([69, 71]\), design characteristics of metasurfaces \([92, 94]\), and of efficacy of light trapping \([7, 129]\) and fluorescent \([100, 101]\) sources). They are also the primary building blocks of quantum and nonlinear phenomena like Förster energy transfer \([169]\), Raman scattering \([73]\), and frequency conversion \([51]\).

A variety of other extension of the formalism should also be possible. In particular, there is an apparent synergy with the work of Angeris, Vučković and Boyd \([39]\) for inverse design applications, as the optimal vectors found using \([15]\) provide intuitive target fields. By combining the respective strengths of both classes of materials, hybrid metal-dielectric structures promise a direction towards better
performing devices. The generalization of (15) to incorporate multiple material regions stands as an aid to these efforts is a current direction of ongoing study.

VI. ACKNOWLEDGMENTS

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VII. APPENDIX

A. Numerical Stability of the Arnoldi Processes.

With perfect numerical accuracy, the convergence of the first column of \( A_f \) is guaranteed to happen in a finite number of iterations. The strictly diagonal elements of each \( V_{f_{i-1}} \) matrix remain constant while the off diagonal coupling coefficients introduced by Sym \( \left[ G_f^0 \right] \) gradually decay with every iterations. Thus, at a certain point, the diagonal \( V_{f_{i-1}} \) eventually overwhelm all of contributions, terminating the \( \Psi_f \) matrix. (For larger susceptibilities more Arnoldi iterations are needed for convergence since the absolute value of \( V_{f_{i-1}} \) elements is smaller.)

Still, there are pitfalls that one may encounter when attempting to numerically implement the Arnoldi iteration for the Green function that stems from the singularity of the outgoing \( N \) waves at the origin. The problem is illustrated by considering the image of \( RN \) under \( G^0 \) [24], with

\[
RN_{\ell,m} = \left( \frac{\ell + 1}{2\ell + 1} \right) (r)^{\ell-1} + O \left( r^{\ell+1} \right) A^{[2]}_{\ell,m} + \left( \frac{\sqrt{\ell(\ell+1)}}{2\ell + 1} \right) (r)^{\ell-1} + O \left( r^{\ell+1} \right) A^{[3]}_{\ell,m} \tag{38}
\]

\[
N_{\ell,m} = \left( \frac{i\ell(\ell-1)!}{r_{f+2} + O \left( r^{-\ell} \right)} \right) A^{[2]}_{\ell,m} + \left( \frac{-i(\ell-1)!\sqrt{\ell(\ell+1)}}{r_{f+2} + O \left( r^{-\ell} \right)} \right) A^{[3]}_{\ell,m} \tag{39}
\]

The image of \( RN_{\ell,m} \) under the Green’s function restricted to a spherical domain with radius \( R \) takes the form

\[
G^0 Rg N = Rg N(r) Rg N_{\ell,m}(r) + O(r) N_{\ell,m}(r) - Rg N(r) A^{[3]}_{\ell,m} \tag{40}
\]

where the final term is the \( \delta \)-function contribution, and the \( Rg N_{\ell,m}(r) \), \( N_{\ell,m}(r) \) terms have forms

\[
N_{\ell,m}(r) = i \int_0^r \int_{\Omega_r} r^2 Rg N(r') \cdot Rg N(r') \ d\Omega' dr' \tag{41}
\]

\[
Rg N_{\ell,m}(r) = i \int_0^r \int_{\Omega_r} r^2 N(r') \cdot Rg N(r') \ d\Omega' dr' \tag{42}
\]

Exploiting the orthogonality of the vector spherical harmonics, simple algebra shows that the leading radial order for \( N_{\ell,m}(r) \) is \( r^{2\ell+1} \) so the term \( N(r) N_{\ell,m}(r) \) has leading radial order \( r^{\ell-1} \), the same as the starting vector \( Rg N(r) \).

At first sight \( Rg N_{\ell,m}(r) \) is more troubling: the leading radial order for \( r^2 N(r) \) is \( r^{-\ell} \) while the leading radial order for \( Rg N(r) \) is \( r^{d-1} \), so it would seem that the integrand has leading radial order \( r^{\ell-1} \), which will result in a \( \log(r) \) term after integration that diverges at the origin. Fortunately, this does not actually happen; a detailed calculation shows that the separate leading order terms from the A^{[2]}_{\ell,m} and A^{[3]}_{\ell,m} contributions cancel out:

\[
Rg N_{\ell,m}(r) = i \int_0^r \left( \frac{i\ell(\ell+1)}{2\ell + 1} r^{\ell-1} A^{[2]}_{\ell,m} \cdot A^{[2]}_{\ell,m} - \frac{i\ell(\ell+1)}{2\ell + 1} r^{\ell-1} A^{[3]}_{\ell,m} \cdot A^{[3]}_{\ell,m} + O \left( r^{d-1} \right) dr' \right) d\Omega = O \left( r^{2\ell} \right) \tag{43}
\]

The key to this cancellation is that the ratio of the leading radial orders of the A^{[2]}_{\ell,m} and A^{[3]}_{\ell,m} terms is \( \sqrt{\ell(\ell+1)}/\ell \). So long as this ratio is intact, the leading radial orders the \( Rg N \) factor do not generate a logarithmic contributions, and in turn this causes the leading order ratio to be maintained under the action of \( G^0 \). Hence, by insuring that this in fact occurs the Arnoldi may continue iterating. Consider any vector

\[
P = p \cdot r^{\ell-1} A^{[2]}_{\ell,m} + \sqrt{\frac{\ell}{\ell+1}} p \cdot r^{\ell-1} A^{[3]}_{\ell,m} \tag{44}
\]

where \( p \) is a constant. (\( Rg N \) is a vector of this form.) The
As anticipated, the vector spherical harmonic components

\[ G^0 = \mathbf{R}_{\ell,m}(r) \mathbf{R}^\dagger_{\ell,m}(r) + N_{\ell,m}(r) N^\dagger_{\ell,m}(r) \]

has components with the same ratio; thus the \( \sqrt{\ell/\ell + 1} \) ratio that prevents a logarithmic term from appearing in \( \mathbf{R}_c^\dagger \) is preserved. By an inductive argument we can extend this to every step of the Arnoldi iteration, with all Arnoldi vectors well-behaved at the origin.

While this is very nice analytically, any numerical implementation of the Arnoldi iteration must take care not to let numerical error push that component ratio away from \( \sqrt{\ell/(\ell + 1)} \) at any step, or the logarithmic divergence will quickly destabilize the entire scheme. Explicitly, the representation of the Arnoldi vectors must maintain the proper asymptotics at the origin. This precludes the use of spatial discretization based representations, since for finite grids discretization error is inevitable and leads to a rapidly growing instability at the origin. For the numerics in this manuscript the radial dependence of the Green’s function and Arnoldi vectors are represented by Taylor series. At larger domain sizes this approach demands a high level of numerical precision, so the Python arbitrary-precision floating-point arithmetic package \texttt{mpmath} \cite{mpmath} is used throughout. When calculating the image of a vector under the Green’s function, the tiny coefficient of \( r^{\ell - 1} \) due to numerical errors from the finite Taylor series and set floating-point arithmetic package \texttt{mpmath} \cite{mpmath} is used throughout. When calculating the image of a vector under the Green’s function, the tiny coefficient of \( r^{\ell - 1} \) due to numerical errors from the finite Taylor series and set floating-point precision is ignored. With sufficiently high floating point precision and Taylor series order the Arnoldi iteration can be performed stably and accurately up to convergence of the \( U \) matrix. Much of the difficulty, and inefficiency, associated with this method stems from working in spherical which are inherently ill defined at the origin.

B. Hessian

For implementations relying on the Hessian we find

\[
\begin{align*}
\frac{\partial^2 q}{\partial \zeta^2} &= \frac{\partial^2 q}{\partial \gamma^2} = \sum_{\ell} \frac{1}{2} \left( \left( S^{(1)}_{\ell} \right) - \left( S^{(4)}_{\ell} \right) \text{Asym} \left[ U_{\ell} \right] \right) A_{\ell,m} \left( \left( S^{(1)}_{\ell} \right) - \text{Asym} \left[ U_{\ell} \right] \right) \\
\frac{\partial^2 q}{\partial \zeta \partial \gamma} &= \sum_{\ell} \Re \left[ \left( S^{(4)}_{\ell} \right) \text{Asym} \left[ U_{\ell} \right] A_{\ell,m} \text{Sym} \left[ U_{\ell} \right] \right] - \Im \left[ \left( S^{(4)}_{\ell} \right) \text{Asym} \left[ U_{\ell} \right] A_{\ell,m} \text{Sym} \left[ U_{\ell} \right] \right] \\
\end{align*}
\]

\[ (50) \]

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