Inverse design of light-matter interactions

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Inverse design represents a paradigm shift in the development of nanophotonic devices, where optimal geometries and materials are discovered by an algorithm rather than symmetry considerations or intuition. Here we present a very general formulation of inverse design that is applicable to atomic interactions in external environments, and derive from this some explicit formulae for optimisation of spontaneous decay rates, Casimir-Polder forces and resonant energy transfer. Using the Q factor of the latter as an example, we use finite-difference time-domain techniques to demonstrate the ability of inverse design algorithms to go far beyond what can be achieved by intuition-based approaches, opening up a new route to their technological exploitation.

Traditional design methods work by specifying a device, then investigating its properties. By contrast, in inverse design the desired property is specified, and an algorithm is left to find a device which fulfils the desired criteria. A naive approach to this would be simply trying all devices that fulfil some set of design constraints. The large space of possible designs renders this numerically unrealistic, meaning that a pre-determined set of designs must be optimised over, at least in the earliest applications of inverse methods to electromagnetic problems\cite{1, 2}. The development of adjoint methods\cite{3} originally used in aerodynamics have made unconstrained inverse design computationally feasible, with the first application used in aerodynamics have made unconstrained inverse design computationally feasible, with the first application in photonics being to low-loss waveguide bends\cite{4}. Adjoint methods were subsequently applied to band gaps\cite{5}, solar cells\cite{6}, on-chip demultiplexers\cite{7} and many more diverse systems — see the recent review articles\cite{8, 9} and references therein.

An area in which inverse design has not yet been applied is virtual-photon mediated interactions, such as Casimir-Polder\cite{10} forces and resonant energy transfer\cite{11}. These processes can be described within a very general formalism known as macroscopic quantum electrodynamics (QED)\cite{12}, where they can be reduced to various functional forms of the classical dyadic Green’s tensor $G$ for a source at $r'$, observation point at $r$ and frequency $\omega$ defined to satisfy

$$\nabla \times \nabla \times G(r, r', \omega) - \frac{\omega^2}{c^2} \varepsilon(r, \omega) G(r, r', \omega) = \Pi (r - r').$$

subject to given boundary conditions. This tensor takes into account both the geometry and material response of an arbitrarily-shaped medium, meaning that an optimal geometry for particular $r, r'$ and $\omega$ is represented by a particular functional form of $G$. It follows that $G$ is the fundamental object which is to be worked with in inverse design of macroscopic QED.

In this article we begin by introducing the underlying formulae for inverse design of light-matter interactions. We then use the specific example of resonant energy transfer combined with finite-difference time domain (FDTD) techniques to demonstrate that the efficiencies achievable in this method are far beyond those found from ‘by-hand’ constructions, opening up a new direction in the design of any light-matter interaction dependent device.

**General formulation.** In order to carry out any optimisation, we need to define a merit function $F$ which we intend to maximise. In traditional presentations of adjoint optimisation, this function is taken to depend on the electromagnetic fields $E, D, B$ and $H$, but all of these are of course deducible from the dyadic Green’s tensor so we consider $F$ as being dependent on only $G(r, r', \omega)$. The merit function should be an observable quantity, so we take it to be a real-valued functional of $G(r, r', \omega)$, integrated over all its arguments:

$$F = \int d^3r \int d^3r' \int_0^\infty d\omega f[G(r, r', \omega)].$$

The integrals allow us to take into account a delocalised source and extended observation volume, as well as multimode effects. The entries of the tensor $G$ are in general complex-valued, so in principle one could consider variations in the real and imaginary parts separately. However, it is more convenient to consider the complex tensors $G$ and $G^*$ as independent, in which case the variation of the merit function with $G$ is;

$$\delta F = 2 \int d^3r \int d^3r' \int_0^\infty d\omega \text{Re} \left[ \frac{\partial f}{\partial G}(r, r', \omega) \circ \delta G(r, r', \omega) \right]$$

where $\circ$ represents the Frobenius product $(A \circ B = A_{ij}B_{ij})$ and $\delta G$ is a change in the Green’s function brought about by an infinitesimal change in the environment. If this change can be considered as being confined to a small volume $V$ containing a number density $n(r'')$ of atoms with polarisabilities $\alpha(r'')$, we can write $G$ in terms of the following Born series;

$$\delta G(r, r', \omega) = \mu_0 \omega^2 \int_V d^3r'' n(r'') \alpha(r'') \times G(r, r'', \omega) \cdot G(r'', r', \omega),$$

where $\delta G = G - G_0$, $G_0$ being the zeroth-order Green’s function, $\mu_0$ is the permeability of free space, $n$ is the number density of atoms, $\alpha$ are the polarisabilities and $G$ is the zeroth-order Green’s function.

The integral in eq. (2) is the expression for the spontaneous decay rate of $G$. It follows that

$$\delta F = \mu_0 \omega^2 \int V d^3r'' n(r'') \alpha(r'') \times \frac{\partial f}{\partial G}(r, r', \omega) \circ G(r, r'', \omega) \cdot G(r'', r', \omega),$$

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which can be considered as being confined to a small volume $V$ containing a number density $n(r'')$ of atoms with polarisabilities $\alpha(r'')$, we can write $G$ in terms of the following Born series;
where \( \mu_0 \) is the vacuum permeability. The change in the merit function is then given by:

\[
\delta F = 2\mu_0 \mathrm{Re} \int d\omega \omega^2 \int d^3r \int d^3r' \int d^3r'' n(r'') \alpha(r'') \\
\times \frac{\partial f}{\partial G}(r, r', \omega) \circ G^T(r', r, \omega) \cdot G(r'', r', \omega),
\]

(5)

where Lorentz reciprocity \( G(r, r', \omega) = G^T(r', r, \omega) \) has been used. Merit functions for observables that depend on \( \nabla G(r, r', \omega) \) can be obtained via the replacements \( G(r, r', \omega) \mapsto \nabla G(r, r', \omega) \) and \( G^T(r'', r', \omega) \mapsto G^T(r'', r', \omega) \nabla \).

There are several features of Eq. (5) worth commenting on. In traditional presentations of adjoint optimisation, the equivalent of (5) is represented as the product of two electric fields. The first is the ‘direct’ field, which is simply the electric field induced by the sources present in the system. The second is the adjoint field, which is that part of the electric field generated by a dipole oscillator at the observation point with an amplitude given by the electric-field derivative of the merit function. The advantage of adjoint methods is that the optimal value of the merit function can be found with only two simulations (rather than a brute force method entailing placement of a dielectric inclusion at each possible point in the optimisation region and repeatedly simulating for each). This is reflected in our version of the merit function change shown in Eq. (5); once the two independent Green’s tensors for a source at \( r' \) and a source at \( r \) in a given environment (e.g. vacuum) have been calculated, \( \delta F \) is known at all points. The link with the adjoint electric field is simply that one of the Green’s tensors in (5) has been transposed.

At this point one has at least two choices for how to practically implement an optimisation — the simplest is an additive scheme illustrated in Fig. 1(i). Here a small block of material is added at the point of maximal \( \delta F \), then the two Green’s tensors in the new geometry are recalculated and combined to find a new optimal position, as shown in Fig. 2.

The second way to implement the optimisation consists of gradually optimising the shape of an initial object by changing its boundary, known as the level-set method [13]. This takes advantage of the fact that Eq. (5) avoids any explicit reference to electric or magnetic fields, thereby avoiding complications with the discontinuities usually found when the fields either side of boundary need to be considered. Here, the initial boundary shape (as well as its subsequent evolution) is encoded by a chosen function \( \phi \). This is defined as negative inside the boundary, zero on it and positive outside, as indicated in Fig. 1(ii). Introducing an artificial ‘time’ parameter \( \delta t \) representing progression along the iterative process, one is led to the following equation of motion governing the shape of the boundary [13]:

\[
\dot{\phi}(r(t), t) + v_n |\nabla \phi(r(t), t)| = 0
\]

(6)

where \( v_n \) is the velocity of motion normal to the surface. Formally, this is an advection equation which can be solved using techniques from fluid dynamics. Taking the volume \( V \) in Eq. (5) to be that defined by the function \( \phi \), we can let:

\[
\int_V d^3r'' \mapsto \int_{\partial V} dA \delta x(r'') = \int_{\partial V} dA v_n \delta t
\]

(7)

where the shape deformation has been assumed to be small, as can be ensured by a sufficiently small time step \( \delta t \) in the evolution process. If the integrand of the \( r'' \) integral in (5) is positive at each iteration, the value of the merit function will continually increase. Positivity of Eq. (5) can then be ensured by using a velocity such that \( \partial F = \int_{\partial V} dA v_n^2 \delta t \), which means identifying:

\[
v_n = 2\mathrm{Re} \int_0^\infty d\omega \int d^3r \int d^3r' \alpha(r'') \\
\times \frac{\partial f}{\partial G}(r, r', \omega) \circ G^T(r'', r, \omega) \cdot G(r'', r', \omega),
\]

(8)

This velocity can be directly calculated for a given \( G \), then inserted into Eq. (6), after which \( \phi \) is evolved for a short time \( \delta t \). This delivers a new \( \phi \), which defines a new geometry, for which we can calculate the new \( G \) and the process iterates. Equation (5) can be directly applied to any quantity that can be expressed in terms of the Green’s dyadic \( G \).
This includes Casimir [14, 15] and Casimir-Polder forces [10, 16], spontaneous decay (Purcell factor) [17, 18], quantum friction [19, 20], interatomic Coulombic decay [21, 22], radiative heat transfer [23, 24], van der Waals forces [25], non-linear optical processes [26] and many more (the latter reference for each of these is where the formula in terms of $\mathbf{G}$ can be found). The merit functions for a selection of these are shown in Fig. 3.

**Example implementation:** In order to demonstrate the application of Eq. (5), we make some simplifying assumptions. We assume that the dielectric additions are homogenous and sufficiently small that the integral over $r''$ can be approximated by the value at its centre $s$:

$$
\delta F = 2\mu_0\alpha n \text{Re} \int d\omega \int d^3r \int d^3r' \times \frac{\partial F}{\partial \mathbf{G}}(r, r', \omega) \odot \mathbf{G}^T(s, r, \omega) \mathbf{G}(s, r', \omega).
$$

(9)

In practice, quantities which depend on the field at a single frequency are considerably more computationally tractable than their multi-frequency counterparts. Here we concentrate on a simple and universal phenomenon which is well-approximated by a single frequency — resonant energy transfer (RET). We will work in the dipole approximation aim to optimise the transfer rate $\Gamma$ between dipole moments $\mathbf{d}_A$ and $\mathbf{d}_D$, meaning we take:

$$
\delta F_{\text{RET}}[\mathbf{G}(r, r', \omega)] = \frac{2\pi \mu_0^2 \omega^4}{\hbar} |\mathbf{d}_A \cdot \mathbf{G}(r, r', \omega) \cdot \mathbf{d}_D|^2 \times \delta(r - r_A)\delta(r' - r_D)\delta(\omega - \omega_D).
$$

(10)

We then have simply:

$$
F_{\text{RET}} = \frac{2\pi \mu_0^2 \omega^4}{\hbar} |\mathbf{d}_A \cdot \mathbf{G}(r_A, r_D, \omega) \cdot \mathbf{d}_D|^2 = \Gamma
$$

(11)

which is the well-known of resonance energy transfer rate $\Gamma$. Using this in Eq. (9), after some algebra one finds

$$
\delta F_{\text{RET}} = \frac{4\pi n \mu_0^2 \omega^4}{\hbar} \text{Re} \left\{ \mathbf{d}_A \mathbf{G}^*(r_A, r_D, \omega) \mathbf{d}_D \times [\mathbf{d}_A \mathbf{G}^T(s, r_A)] \cdot [\mathbf{G}(s, r_D) \mathbf{d}_D] \right\}
$$

(12)

which is the basic equation we will work with for the rest of this article.

In order to demonstrate the main features of the method we restrict ourselves to systems with translational invariance along one axis, meaning they can be considered as effectively two-dimensional. In order to validate the two-dimensional RET results that we will calculate (as well as the general FDTD approach), it is necessary to have an analytic expression for RET in two dimensions. To our knowledge this does not appear in the literature, so we shall present a new expression here. Formally, 2D-RET is equivalent to taking a pair of ‘line dipoles’ each consisting of two infinitely extended parallel oppositely-charged wires in three dimensions, as discussed in detail in [27]. The relevant Green’s tensor is:

$$
\mathbf{G}_{2D}(r, r', \omega) = \frac{i}{4} \left( \mathbb{I} + \frac{\nabla \otimes \nabla}{\omega^2/c^2} \right) H_0^{(1)}(k_{||}\rho) e^{ik_z z}
$$

(13)

where $H_n^{(1)}$ is the Hankel function of the first kind. Using this in the general formula for RET with donor-acceptor separation $\rho$ and transition frequency $\omega$. This can be directly substituted into Eq. (11) and the derivatives evaluated. The result is a lengthy expression, which can be simplified by noting that in situations of practical interest the dipoles are often randomly oriented necessitating an isotropic average, which gives:

$$
\Gamma_{\text{iso}}^{2D} = \frac{2\pi \mu_0^2 \omega^4}{\hbar} \frac{1}{16\zeta} \left\{ 2\zeta H_0^{(1)}(\zeta) - H_1^{(1)}(\zeta) \right\} H_0^{(2)}(\zeta) + H_2^{(1)}(\zeta) H_1^{(2)}(\zeta)
$$

(14)

where $H_n^{(1)}$ and $H_n^{(2)}$ are Hankel functions of the first and second kind respectively, and $\zeta = \omega_D\rho/c$. As an aside, we note that the 2D-RET expression (14) is qualitatively different from its three-dimensional counterparts, which is most apparent in asymptotic limits. At short distances (much less than the wavelength), 3D-RET falls off as $1/\rho^6$, whereas in 2D this changes to $1/\rho^4$. At long distances, the three-dimensional result decays as $1/\rho^2$, whereas the 2D result here has a $1/\rho$ behaviour. The expression above is used to validate the general FDTD approach to 2D-RET, with detailed discussion in the Appendix.

We can now calculate the effect of arbitrary 2D geometries on RET by examining the dimensionless quality factor $Q = \Gamma/\Gamma_0$, where $\Gamma_0$ is the rate in vacuum. We initially choose some geometries which are expected to enhance RET, these are shown in Fig. 4, and give a maximum $Q$ in the low hundreds.

We can now assess if the iterative optimisation techniques can improve on the examples chosen by hand. We use the additive approach shown in Fig. 1(i), the results of which are shown in Fig. 5. An extremely high $Q$ is found, reaching approximately $10^5$ after 250 iterations — orders of magnitude higher than any enhancement found in the traditional designs shown in Fig. 4. It is worth noting that this extraordinarily high $Q$ is achieved with a much smaller amount of dielectric material than in the traditional designs, even though the dielectric constant is identical.

In this article we have presented a convenient and system-agnostic version of adjoint optimisation of electromagnetism based entirely on the electromagnetic dyadic Green’s tensor. This allows the techniques of inverse design to be applied to any of the vast number of interactions and processes which can be expressed in terms of this tensor. As an example we chose resonant energy transfer in two dimensions, showing orders of magnitude improvement in engineering potential compared to hand-made designs, while also deriving some new analytic results along the way. Extensions of our work could include application of the general level-set optimisation equation presented here, three-dimensional simulations and consideration of other observables including for example quantum yield of fluorescence processes. Inverse
design of solar cells [6] can now be conducted at a microscopical level by explicitly optimising each step in the energy transport chain, leading to large potential increases in efficiency.

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Appendix A: Computational approach

Since the Green’s tensor appearing in the main text is only known analytically in the very simplest of geometries, we will in general need evaluate it numerically. There are a variety of ways of doing this, one is to use a finite difference time domain (FDTD) solver. Here we will make use of the free and open-source MEEP FDTD library [28].

The $ij$ component of the Green’s tensor $G$ describes the $i$ component of the electric field at $r$ coming from the $j$ component of a current source at $r_s$. For a point source at $r_s$, we have:

$$
E(r, \omega) = \frac{1}{i\mu_0 \omega} G(r, r_s, \omega) j(\omega)
$$

(A1)

In FDTD the source is of course specified in the time domain, necessitating an additional Fourier transform.
Given a suitable \( \mathbf{j}(t) \) the resulting \( \mathbf{E} \) can be found directly by any FDTD software, giving us everything we need to deduce \( \mathbf{G} \). Similarly to [29], the current source used in the simulations here is a short Gaussian pulse (the built-in Meep function \texttt{GaussianSource}) polarised in the \( j \) direction,

\[
\mathbf{j}(t) = A \exp \left[ -\frac{(t - t_0)^2}{2w^2} \right] \mathbf{e}_j
\]  

where \( A \) is an arbitrary amplitude [appearing on both sides of Eq. (A1)], \( t_0 \) is the time at which the maximum is reached and \( w \) is the width. The Fourier transform of this is:

\[
\mathbf{j}(\omega) = \frac{A}{\Delta f} \exp \left[ i\omega t_0 - \frac{(\omega - \omega_0)^2}{2\Delta f^2} \right] \mathbf{e}_k
\]

and the simulation is allowed to run for long enough that all fields have decayed away, which is taken here as being 100 times the temporal width of the Gaussian. The Fourier-transformed electric fields are found from the time domains fields which in turn are found by direct simulation. Dividing these fields by \( i\omega \mathbf{e}_j(\omega) \), one row of the Green’s tensor is known, corresponding to a particular source polarisation direction \( k \). This is done (in parallel) for all three source polarisations, giving us all nine components of \( \mathbf{G} \). In most situations the Green’s tensor is symmetric so that only six of these components are independent, allowing further increases in efficiency.

Comparing this with the results coming out of the FDTD approach, we find excellent agreement as shown in Fig. 6.
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