Inverse design of light–matter interactions in macroscopic QED

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Abstract
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 Purcell enhancement of the latter as a simple example, we employ finite-difference time-domain techniques in a proof-of-principle demonstration of our formula, finding enhancement of the rate many orders of magnitude larger than a selection of traditional designs.

1. Introduction

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 naïve 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 predetermined set of designs must be optimised over, at least in the earliest applications of inverse methods to electromagnetic problems [1, 2]. The development of adjoint methods [3] originally used in aerodynamics have made unconstrained inverse design computationally feasible, with the first application in photonics being to low-loss waveguide bends [4]. Adjoint methods were subsequently applied to band gaps [5], solar cells [6], on-chip demultiplexers [7] and many more diverse systems—see the recent review articles [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 [10] forces and resonant energy transfer [11]. These processes can be described within a very general formalism known as macroscopic quantum electrodynamics (QED) [12], where they can be reduced to various functionals of the classical dyadic Green’s tensor \( G \) for a source at \( r' \), observation point at \( r \) and frequency \( \omega \) defined to satisfy

\[
\nabla \times \nabla \times G(r, r', \omega) - \frac{\omega^2}{c^2} \varepsilon(r, \omega) G(r, r', \omega) = \delta(r - r')
\]  

subject to given boundary conditions. Though \( G \) is a completely classical quantity, it is a vital ingredient for the description of quantised electromagnetic fields in media [12, 13], and thereby quantum light–matter interactions in their vicinity. It appears, for example, in the coupling constants found in master equations governing weak light–matter interactions (e.g. those for atomic waveguide QED [14]), as well as in matrix elements in the strong coupling regime (e.g. references [15–17]). The quantised electric field in a region with permittivity \( \varepsilon(r, \omega) \) and unit permeability is given via \( G \) as [12],

\[
\hat{E}(r) = i \int_0^\infty d\omega \int d^3r' \frac{\omega^2}{c^2} \sqrt{\frac{\hbar}{\pi \varepsilon_0}} \text{Im} \varepsilon(r', \omega) G(r, r', \omega) \cdot \hat{b}(r', \omega) + h.c.
\]  

where \( \hat{b}^\dagger, \hat{b} \) are a set of bosonic creation and annihilation operators for the medium-assisted quantised
electromagnetic field\(^4\). The Green’s tensor \(\mathbf{G}\) takes into account both the geometry and material response of an arbitrarily-shaped medium, meaning that an optimal geometry for particular \(\mathbf{r}, \mathbf{r}'\) and \(\omega\) is represented by a particular functional form of \(\mathbf{G}\). It follows that \(\mathbf{G}\) is the fundamental object which is to be worked with in inverse design of macroscopic QED.

In this article we begin in section 2 by introducing the underlying formulae for inverse design of light–matter interactions. In section 3 we 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 and optimisation of a broad class of light–matter interactions. We conclude with some discussion and suggestions for further work in section 4.

2. 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 \(\mathbf{E}, \mathbf{D}, \mathbf{B}\) and \(\mathbf{H}\), but all of these are of course deducible from the dyadic Green’s tensor so we consider \(F\) as being dependent on only \(\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)\). The merit function should be an observable quantity, so we take it to be a real-valued functional of \(\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)\), integrated over all its arguments:

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

The integrals allow us to take into account a delocalised source and extended observation volume, as well as multi-mode effects. The entries of the tensor \(\mathbf{G}\) 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 \(\mathbf{G}\) and \(\mathbf{G}^*\) as independent, in which case the variation of the merit function with \(\mathbf{G}\) is;

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

where \(\circ\) represents the Frobenius product \((A \circ B = \sum_i \sum_j A_{ij} B_{ij})\) and \(\delta \mathbf{G}\) 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 \(n(\mathbf{r}')\) of atoms with polarisabilities \(\alpha(\mathbf{r}')\), as shown in appendix A we can write \(\mathbf{G}\) in terms of the following Born series:

\[
\delta \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \mu_0 \omega^2 \int_V d^3r'' n(\mathbf{r}'') \alpha(\mathbf{r}'') \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{G}(\mathbf{r}'', \mathbf{r}', \omega),
\]

where \(\mu_0\) is the vacuum permeability. The change in the merit function due to an additional small piece of dielectric material is then given by;

\[
\delta F = 2 \mu_0 \omega^2 \int d\omega \int d^3r \int d^3r' \int_0^\infty d\omega' n(\mathbf{r}'') \alpha(\mathbf{r}'') \frac{\partial f}{\partial \mathbf{G}} (\mathbf{r}, \mathbf{r}', \omega) \circ \mathbf{G}^T(\mathbf{r}'', \mathbf{r}, \omega) \cdot \mathbf{G}(\mathbf{r}'', \mathbf{r}', \omega),
\]

where Lorentz reciprocity \(\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{G}^T(\mathbf{r}', \mathbf{r}, \omega)\) has been used. Merit functions for observables that depend on \(\nabla_i \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)\) can be obtained via the replacements \(\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \mapsto \nabla_i \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)\) and \(\mathbf{G}^T(\mathbf{r}', \mathbf{r}, \omega) \mapsto \mathbf{G}^T(\mathbf{r}', \mathbf{r}, \omega) \nabla_i\), with the most general form being a sum over all such derivative contributions.

Equation (6) is our main result, representing a formula of remarkable power and utility, ready for direct application a variety of processes. Before discussing some particular cases, there are several features of equation (6) worth commenting on. In traditional presentations of adjoint optimisation, the equivalent of equation (6) is expressed 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 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 our version of the merit function change shown in (6); once the two independent Green’s

\(^4\) These are usually called \(f\) and \(\bar{f}\) in macroscopic QED, here we avoid that notation in order to avoid confusion with the merit functions \(f\) introduced later.
and shape of the added blocks can be adjusted to take into account constraints from a given additive manufacturing process. Here, the initial boundary shape (as well as its subsequent evolution) is encoded by a chosen function \( \phi \) representing iteration, one is led to the following equation of motion governing the shape of the boundary deformation:

\[
\frac{\partial \phi}{\partial t} = \mathbf{v}_n \cdot \nabla \phi - \nabla \cdot \mathbf{F}.
\]

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 [36]. Here, the initial boundary shape (as manufactured) is transposed by taking advantage of Lorentz reciprocity. This includes Casimir [18, 19] and Casimir–Polder forces [10, 20], spontaneous decay (Purcell factor) [21, 22], quantum friction [23, 24], interatomic Coulombic decay [25, 26], radiative heat transfer [27, 28], van der Waals forces [29], non-linear optical processes [30, 31], environment-induced coherence [32, 33], resonant energy transfer [11, 14] and many more (the latter reference for each of these is where the formula in terms of the Green’s tensor can be found). The merit functions for a selection of these are shown in Table 1.

| Observable          | Merit function integrand \( f \) | Merit function change \( \delta F \) |
|---------------------|-----------------------------------|--------------------------------------|
| Spontaneous decay   | \( \frac{2\mu}{h} \frac{d^2}{d\omega^2} \mathbf{d}_k \cdot \Im G(r, r', \omega) \cdot \mathbf{d}_k \times 4\pi \omega | \frac{4\pi}{\omega} \Im \left\{ \mathbf{d}_k \cdot G^\dagger(s, r_k, \omega_0) \cdot \mathbf{G}(s, r_k, \omega_0) \cdot \mathbf{d}_k \right\} |
| Casimir–Polder force| \( \frac{2}{\omega} \sum_{\omega} \frac{d\omega^2}{d\omega^2} \mathbf{d}_k \cdot \left[ \nabla G(r, r', \omega) \right] \cdot \mathbf{d}_k \times 4\pi \omega | \frac{4\pi}{\omega} \Re \left\{ \mathbf{d}_k \cdot G(s, r_k, \omega_0) \cdot \mathbf{d}_k \times \left[ \mathbf{d}_k \cdot G^\dagger(s, r_k, \omega_0) \right] \right\} |
| Resonance energy    | \( \frac{2\mu}{h} \frac{d^2}{d\omega^2} \mathbf{d}_k \cdot \mathbf{G}(r, r', \omega) \cdot \mathbf{d}_k \times 4\pi \omega | \frac{4\pi}{\omega} \Im \left\{ \mathbf{d}_k \cdot G^\dagger(s, r_k, \omega_0) \cdot \mathbf{G}(s, r_k, \omega_0) \cdot \mathbf{d}_k \right\} |

By choosing the adjoint electric field \( \mathbf{E}^* \) such that the following integral in (6) is positive at each iteration, the value of the merit function will continually increase. Positivity of (6) can then be ensured by using a velocity such that \( \mathbf{v}_n = 2 \Re \int_0^\infty d\omega \int d^3r \int d^3r' \alpha(r') \frac{\partial F}{\partial G} (r, r', \omega) \odot G^\dagger(r', r, \omega) \cdot \mathbf{G}^\dagger(r', r, \omega). \)
This velocity can be directly calculated for a given $G$, then inserted into the advection equation, after which $\phi$ is evolved for $\delta t$. This delivers a new $\phi$, which defines a new geometry, for which the new $G$ can be calculated and the process iterates. Complex manufacturing constraints such elimination of highly curved regions, gaps, or ‘bridges’ can be enforced within in the level set method, as discussed in detail in reference [37].

3. Example implementation

In order to succinctly demonstrate the use of (6), 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 $s$:

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

(8)

In practice, quantities which depend on the field at a single frequency and at a single position are considerably more computationally tractable than their multi-frequency, bulk medium counterparts. Here we concentrate on a simple and universal phenomenon which is well-approximated by radiation of a single dipole (casimir–polder forces) would require recalculation of the Green’s tensors, but calculations of quantities dependent on a continuous spectrum (e.g. ground-state Casimir–Polder forces) would require re-calculation of $G$ at very many frequencies.

We will work in the dipole approximation and aim to optimise the RET rate $\Gamma$ between dipole moments $d_A$ and $d_D$, meaning we take [34]:

$$F_{\text{RET}}[G(r, r', \omega)] = \frac{2\pi \mu_0^2 \omega^4}{c} |d_A \cdot G(r, r', \omega) \cdot d_D|^2 \delta(r - r_A)\delta(r' - r_D)\delta(\omega - \omega_D).$$

(9)

We then have simply:

$$F_{\text{RET}} = \frac{2\pi \mu_0^2 \omega^4}{c} |d_A \cdot G(r_A, r_D, \omega_D) \cdot d_D|^2 = \Gamma$$

(10)

which is the well-known expression of the resonance energy transfer rate $\Gamma$. Using this in (8), after some algebra one finds

$$\delta F_{\text{RET}} = \frac{4\pi \mu_0^2 \omega^4}{c} \Re \{d_A \cdot G^*(r_A, r_D, \omega_D) \cdot d_D |d_A \cdot G^T(s, r_A, \omega_D)| \cdot |G(s, r_D, \omega_D) \cdot d_D|\}$$

(11)

which is the equation we will work with from here on.

To simplify our presentation of the main features of the method we restrict ourselves to systems with translational invariance along one axis, in other words those which 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. 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 reference [38]. The Green’s tensor from [38] can be directly substituted into (10), resulting in 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 (see appendix B), which gives:

$$\Gamma_{\text{iso}} = \frac{2\pi \mu_0^2 \omega^4}{c} d_A^2 d_D^2 \frac{1}{16\pi} \left\{ \left[ 2 \zeta H_0^{(1)}(\zeta) - H_1^{(3)}(\zeta) \right] H_0^{(2)}(\zeta) + H_1^{(1)}(\zeta) H_2^{(2)}(\zeta) \right\}$$

(12)

where $H_n^{(1)}$ and $H_n^{(2)}$ are Hankel functions of the first and second kind respectively, $\zeta = \omega_D \rho / c$, $\rho = |r_A - r_D|$, $d_A = |d_A|$ and $d_D = |d_D|$. The Green’s tensor from [38] can also be substituted into equation (11) to give an initial indication of where material should be placed in order to optimise the RET process, this is shown in figure 2.

Equation (12) can be used to validate our general numerical approach, in which we used the free FDTD software Meep [39] to calculate the Green’s tensors entering (10) and (11). In order to do this we note that the $ij$ component of the Green’s tensor $G(r, r', \omega)$ describes the $i$th component of an electric field at observed at $r$ due to the $j$ component of a current source at $r'$. 

$$\mathbf{E}(r, \omega) = i \mu_0 \omega \int d^3 r' G(r, r', \omega) \cdot j(r', \omega).$$

(13)
Figure 2. Merit function gradient (11) using the Green’s tensor from [38] and dipoles aligned along the x axis with donor transition wavelength of $\pi \mu m$. Only the functional form and sign (not the magnitude) of the merit function gradient are of relevance to an optimisation, so the values here are shown normalised to the magnitude of $\delta F$ at the midpoint between the two dipoles.

Figure 3. Numerical vs analytic results for 2D-RET with donor and acceptor dipole moments parallel and of identical magnitude, with a transition wavelength of 500 nm. Four different resolutions are shown, and in all cases agreement with (12) is excellent until the interatomic distance approaches the pixel size used in each simulation, indicated by the the vertical dashed lines. The simulations shown in figures 4 and 5 are all carried out at a resolution of 0.1 pixel/$\mu m$ and are constrained to only allow material to be placed at least 1 $\mu m$ from either donor or acceptor. This is far above the distances at which the discretisation has an effect, as indicated in by the solid vertical line.

For a point source at $r_s$

$$j(r', \omega) = j(\omega)\delta(r' - r_s) \quad (14)$$

so equation (13) becomes

$$E(r, \omega) = i\mu_0 \omega G(r, r_s, \omega) \cdot j(\omega) \quad (15)$$

where $j(\omega) \equiv j(r_s, \omega)$ is the source current in the frequency domain. The FDTD implementation found in Meep (or in other FDTD tools) can calculate the electric field of a time-domain source $j(t)$, which gives us everything we need to deduce $G$. To do this, we introduce a current source in a similar way to reference [40], namely a short Gaussian pulse (the built-in Meep function Gaussian source) polarised in the $j$ direction. The corresponding time-domain current is given by;

$$j(t) = A \exp \left[ i\omega t - \frac{(t - t_0)^2}{2\Delta f^2} \right] e_j \quad (16)$$

where $A$ is an arbitrary amplitude [appearing on both sides of equation (15)], $t_0$ is the time at which the maximum is reached and $\Delta f$ is the temporal width of the Gaussian. The Fourier transform of this is;

$$j(\omega) = \frac{A}{\Delta f} \exp \left[ i\omega t_0 - \frac{(\omega - \omega_0)^2}{2\Delta f^2} \right] e_j \quad (17)$$
The time-domain 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 (no change is observed in the results when the simulation time is varied, as long as it is more than roughly 10 times with width of the Gaussian). The result is a set of time-domain electric fields, which are then Fourier-transformed. Dividing these transformed fields component-wise by $i \omega \varepsilon_0 \mu_0 \omega j(\omega)$, one row of the Green’s tensor is obtained, corresponding to a particular source polarisation direction $j$. This can be done in parallel for all three source polarisations, giving all nine components of $G$. In most situations the Green’s tensor is symmetric so that only six of these components are independent, allowing further increases in efficiency. The RET rate obtained by calculating the FDTD Green’s tensor in this way and substituting it into equation (10) is compared with the analytic
expression (12) in figure 3. Close agreement is found until separations are comparable to pixel size, as is to be expected in any numerical scheme.

We can now calculate the effect of arbitrary 2D geometries on RET. To do this we examine an analogue of the well-known Purcell factor for spontaneous decay [21], by investigating a Purcell-type factor \( F_p = \Gamma / \Gamma_0 \), where \( \Gamma_0 \) is the RET rate in vacuum. Broadly analogous quantities for laser cavities have previously been optimised via algorithmic design (e.g. Q factors in [41]), but without coupling to atoms and without the power and flexibility of the Green’s tensor approach. We begin by choosing some geometries which are expected to enhance RET, these are shown in figure 4, giving a maximum \( F_p \) in the hundreds. Iterative optimisation techniques can dramatically improve on this performance. In order to demonstrate this, we use the additive approach shown in figure 1, the results of which are shown in figure 5. An extremely large enhancement is found, reaching a factor of approximately 10^5 after 250 iterations—orders of magnitude higher than any enhancement found in the traditional designs shown in figure 4. It is worth noting that this extraordinarily high enhancement is achieved with a much smaller amount of dielectric material than in the traditional designs, even though the dielectric constant is identical. While a full eigenmode analysis of the structures shown in figure 5 is not the focus of the current work, some sense can be made of the apparently chaotic-looking designs by noting the waveguide-like structures in the regions into which dipole radiation is strongest (perpendicularly to the dipole moment). These consist of thin lines of dielectric material spaced approximately half a wavelength apart, acting in some sense like highly-reflective Bragg mirrors for the excitations travelling from donor to acceptor. Finally we note that while all our results are scale-invariant (depending only on the ratio of the transition wavelength to the interatomic separation), the minimum feature size in our simulations is 0.1 \( \mu m \)—this is broadly consistent with the level to which complex structures can be manufactured (see, e.g. [43]).

4. Conclusions

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 [19, 20, 22, 24, 26, 28, 29, 31, 33]. As a simple example we chose resonant energy transfer in two dimensions, showing orders of magnitude improvement in engineering potential compared to hand-made designs. The methods of inverse design presented here could also be used to investigate exceptional points in parameter space found by minimising some distance functional. One major extension of our work would be to non-reciprocal media, which would be taken into account by simply by leaving the Green’s tensors in equation (6) untransposed, although the resulting simulations would no longer necessarily be able to take advantage of the speedup provided by the adjoint method. Further extensions of our work will include exploitation 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 and environment-induced coherence.

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Appendix A. The Born series for the Green’s tensor

The Green’s tensor is defined to satisfy equation (1), which we rewrite here for a Green’s tensor \( \tilde{G} \) associated with a permittivity \( \varepsilon(r, \omega) \)

\[
\nabla \times \nabla \times \tilde{G}(r, r', \omega) = \frac{\omega^2}{c^2} \tilde{\varepsilon}(r, \omega) \tilde{G}(r, r', \omega) = \delta(r - r').
\]

Following [44], we assume that the permittivity \( \varepsilon(r, \omega) \) can be decomposed into ‘background’ permittivity \( \varepsilon_0(r, \omega) \) for which the Green’s tensor \( \tilde{G}_0 \) is known, plus a small correction \( \chi(r, \omega) \), meaning that we have;

\[
\varepsilon(r, \omega) = \varepsilon_0(r, \omega) + \chi(r, \omega)
\]
and
\[ \nabla \times \nabla \times G(r, r', \omega) - \frac{\omega^2}{c^2} \varepsilon(r, \omega) G(r, r', \omega) = \mathbb{1} \delta (r - r'). \quad (A.3) \]

Equation (A.2) can be used to rearrange equation (A.1) to;
\[ \nabla \times \nabla \times G(r, r', \omega) - \frac{\omega^2}{c^2} \varepsilon(r, \omega) G(r, r', \omega) = \mathbb{1} \delta (r - r') + \frac{\omega^2}{c^2} \chi(r, \omega) G(r, r', \omega). \quad (A.4) \]

The formal solution to this equation can be written as a Born (or Dyson) series;
\[ \bar{G}(r, r', \omega) = G(r, r', \omega) + \frac{\omega^2}{c^2} \int_V d^3 s \chi(s, \omega) G(r, s, \omega) \cdot \bar{G}(s, r', \omega). \quad (A.5) \]

where the integral runs over the volume V of the dielectric perturbation. The formal solution (A.5) can be verified by direct substitution back into equation (A.4) and use of the fact that \( G(r, r', \omega) \) satisfies equation (A.3). Equation (A.5) is exact but infinitely recursive. Nevertheless, by repeated use of (A.5) one can obtain a perturbative solution for \( G(r, r', \omega) \) in powers of \( \chi \). The first term in the series is;
\[ G(r, r', \omega) = G(r, r', \omega) + \frac{\omega^2}{c^2} \int_V d^3 s \chi(s, \omega) G(r, s, \omega) \cdot \bar{G}(s, r', \omega). \quad (A.6) \]

If the background medium is vacuum, then the permittivity change \( \chi(r, \omega) \) is
\[ \chi(r, \omega) = \frac{n(r) \alpha(r)}{\varepsilon_0}. \quad (A.7) \]

which means it is equal to the susceptibility, and therefore can be converted to a polarisability \( \alpha \) using the Clausius–Mosotti law in the dilute limit [45]:
\[ \chi(r, \omega) = n(r) \alpha(r)/\varepsilon_0. \quad (A.8) \]

Here \( n(r) \) is the number density of atoms with polarisability \( \alpha \) in the perturbing object, and \( \varepsilon_0 = 1/(\mu_0 c^2) \) is the permittivity of free space. Identifying \( \delta G = \bar{G} - G \), one is left with;
\[ \delta G(r, r', \omega) = \mu_0 \omega^2 \int_V d^3 s n(s) \alpha(s) G(r, s, \omega) \cdot G(s, r', \omega), \quad (A.9) \]

which is equation (5) in the main text.

### Appendix B. Resonance energy transfer with translational invariance

The rate of resonance energy transfer for atoms with real transition dipole moments \( \mathbf{d}_D \) and \( \mathbf{d}_A \) is given by equation (10), which can be equivalently written as;
\[ \Gamma = \frac{2\pi \mu_0^2 \omega_0^4}{h} \text{Tr} \left[ \mathbf{d}_A \otimes \mathbf{d}_A \cdot G(r_A, r_D, \omega_D) \cdot \mathbf{d}_D \otimes \mathbf{d}_D \cdot G^* (r_D, r_A, \omega_D) \right]. \quad (B.1) \]

An average over random orientations of donor and acceptor can be taken by letting (see, for example, equation (16) in [46]);
\[ \mathbf{d}_D \otimes \mathbf{d}_D \to \frac{1}{3} \mathbf{d}_D^2 \quad \mathbf{d}_A \otimes \mathbf{d}_A \to \frac{1}{3} \mathbf{d}_A^2 \quad (B.2) \]

where \( d_A = |\mathbf{d}_A| \) and \( d_D = |\mathbf{d}_D| \). Under these conditions equation (B.1) becomes;
\[ \Gamma_{iso} = \frac{2\pi \mu_0^2 \omega_0^4}{h} \text{Tr} \left[ G(r_A, r_D, \omega_D) \cdot G^*(r_D, r_A, \omega_D) \right]. \quad (B.3) \]

The Green’s tensor for a system translationally invariant along the z direction is taken directly from [38], reading;
\[ G(r, r', \omega) = \frac{i}{4\xi} \text{diag} \left[ H_1^{(1)}(\xi), k\rho H_0^{(1)}(\xi) - H_1^{(1)}(\xi), k\xi H_0^{(1)}(\xi) \right] \quad (B.4) \]

where \( H_0^{(1)} \) is the Hankel function of the first kind and \( \xi = \omega_D/\rho/c \) with \( \rho = |r - r'| \). Substituting this into (B.3) and using
\[ H_0^{(1)}(x)^* = H_0^{(1)}(x) \quad (B.5) \]

where \( H_0^{(2)} \) is the Hankel function of the second kind, one arrives directly at equation (12) in the main text.
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References

[1] Spuhler M M, Offrein B J, Bona G-L, Germann R, Massarek I and Erni D 1998 A very short planar silica spot-size converter using a nonperiodic segmented waveguide J. Lightwave Technol. 16 1680–5

[2] Dobson D C and Cox S J 1999 Maximizing band gaps in two-dimensional photonic crystals SIAM J. Appl. Math. 59 2108–20

[3] Jameson A 1989 Aerodynamic design via control theory J. Sci. Comput. 3 233–60

[4] Jensen J S and Sigmund O 2004 Systematic design of photonic crystal structures using topology optimization: low-loss waveguide bends Appl. Phys. Lett. 84 2022–4

[5] Kao C Y, Osher S and Yablonovitch E 2005 Maximizing band gaps in two-dimensional photonic crystals by using level set methods Appl. Phys. B 81 235–44

[6] Ganapati V, Miller O D and Yablonovitch E 2014 Light trapping textures designed by electromagnetic optimization for subwavelength thick solar cells IEEE J. Photovolt. 4 175–82

[7] Piggott A Y, Lu J, Lagoudakis K G, Jan P, Babinec T M and Vucković J 2015 Inverse design and demonstration of a compact and broadband on-chip wavelength demultiplexer Nat. Photon. 9 374–7

[8] Jensen J S and Sigmund O 2011 Topology optimization for nano-photronics Laser Photon. Rev. 5 308–21

[9] Molesky S, Lin Z, Piggott A Y, Jin W, Vucković J and Rodriguez A W 2018 Inverse design in nanophotonics Nat. Photon. 12 659–70

[10] Casimir H B G and Polder D 1948 The influence of retardation on the London–van der Waals forces Phys. Rev. 73 360–72

[11] Förster T 1948 Zwischenmolekulare energiewanderung und fluoreszenz Ann. Phys., Lpz. 437 55–75

[12] Gruner T and Welsch D-G 1996 Green-function approach to the radiation-field quantization for homogeneous and inhomogenous Kramers–Kronig dielectrics Phys. Rev. A 53 1816–29

[13] Huttner B and Barnett S M 1992 Quantization of the electromagnetic field in dielectrics Phys. Rev. A 46 4306–22

[14] Asenjo-Garcia A, Moreno-Cardoner M, Albrecht A, Kimble H J and Chang D E 2017 Exponential improvement in photon storage fidelities using subradiance & ‘selective radiance’ in atomic arrays Phys. Rev. X 7 031024

[15] Buhmann S Y and Welsch D-G 2008 Casimir–Polder forces on excited atoms in the strong atom-field coupling regime Phys. Rev. A 77 012110

[16] Esfandarpour S, Hassan S, Bennett R and Buhmann S Y 2018 Cavity-QED interactions of two correlated atoms J. Phys. B: At. Mol. Opt. Phys. 51 094004

[17] Lentrodt D and Evers J 2020

[18] Klatt J, Belc V, Malyk I, Albrecht A, Kimble H J and Chang D E 2019 Cavity-QED interactions of two correlated atoms J. Phys. B: At. Mol. Opt. Phys. 51 094004

[19] Raabe C, Knoll L and Welsch D-G 2007 Three-dimensional Casimir force between absorbing multilayer dielectrics Phys. Rev. A 68 033810

[20] Buhmann S Y, Knoll L, Welsch D-G and Dung H T 2004 Casimir–Polder forces: a nonperturbative approach Phys. Rev. A 70 052117

[21] Purcell E M 1946 Spontaneous emission probabilities at radio frequencies Bull. Am. Phys. Soc. 69 674

[22] Jouliam K, Carminati R, Muler J-P and Grefett J I 2003 Deﬁnition and measurement of the local density of electromagnetic states close to an interface Phys. Rev. B 68 245405

[23] Penry J B 1997 Shearing the vacuum–quantum friction J. Phys.: Condens. Matter. 9 10301–20

[24] Klatt J, Belén Farias M, Dalvit D A R and Buhmann S Y 2017 Quantum friction in arbitrarily directed motion Phys. Rev. A 95 052310

[25] Cederbaum L S, Zobeley J and Tarantelli F 1997 Giant intermolecular decay and fragmentation of clusters Phys. Rev. Lett. 79 4778–81

[26] Hemmerich J L, Bennett R and Buhmann S Y 2018 The influence of retardation and dielectric environments on interatomic Coulombic decay Nat. Commun. 9 2934

[27] Polder D and Van Hove M 1971 Theory of radiative heat transfer between closely spaced bodies Phys. Rev. B 4 3303–14

[28] Volokitin A I and Persson B N J 2001 Radiative heat transfer between nanostructures Phys. Rev. B 65 205404

[29] Buhmann S Y, Dung H T and Welsch D-G 2004 The van der Waals energy of atomic systems near absorbing and dispersing bodies J. Opt. B: Quantum Semiclass. Opt. 6 5127–35

[30] Poddubny A N, Iorsh I V and Sukhorukov A A 2016 Generation of photon–plasmon quantum states in nonlinear hyperbolic metamaterials Phys. Rev. Lett. 117 123901

[31] Lindel F, Bennett R and Buhmann S Y 2019 Probing the sculpted quantum vacuum: quantum optics of nonlinear crystals (arXiv:1905.10200[quant-ph])

[32] Agarwal G S 2000 Anisotropic vacuum–induced interference in decay channels Phys. Rev. Lett. 84 5500–3

[33] Lassalle E, Lalanne P, Aljunid S, Genevet P, Stout B, Durt T and Willkowski D 2020 Long-lifetime coherence in a quantum emitter induced by a metasurfaces Phys. Rev. A 101 013837

[34] Dung H T, Knoll L and Welsch D-G 2002 Intermolecular energy transfer in the presence of dispersing and absorbing media Phys. Rev. A 65 043813

[35] Liang X and Johnson S G 2013 Formulation for scalable optimization of microcavities via the frequency-averaged local density of states Opt. Express 21 30812

[36] Osler S and Sethian J A 1988 Fronts propagating with curvature-dependent speed: algorithms based on Hamilton–Jacobi formulations J. Comput. Phys. 79 12–49

[37] Piggott A Y, Petykiewicz J, Lu J and Vuckovic J 2017 Fabrication-constrained nanophotonic inverse design Sci. Rep. 7 1–7

[38] Martin O J and Pillier N B 1998 Electromagnetic scattering in polarizable backgrounds Phys. Rev. E 58 3909–15

[39] Oskooi A F, Roundy D, Ibanescu M, Bermel P, Joannopoulos J D and Johnson S G 2010 MEEP: a flexible free-software package for electromagnetic simulations by the FDTD method Comput. Phys. Commun. 181 687–702

[40] Ding W, Hsu L-Y and Schatz G C 2017 Plasmon-coupled resonance energy transfer: a real-time electrodynamics approach J. Chem. Phys. 146 064109
[41] Frei W R, Johnson H T and Choquette K D 2008 Optimization of a single defect photonic crystal laser cavity J. Appl. Phys. 103 033102
[42] Berenger J P 1994 A perfectly matched layer for the absorption of electromagnetic waves J. Comput. Phys. 114 185–200
[43] Su L, Piggott A Y, Sapra N V, Petykiewicz J and Vučković J 2018 Inverse design and demonstration of a compact on-chip narrowband three-channel wavelength demultiplexer ACS Photonics 5 301–5
[44] Buhmann S Y and Welsch D-G 2005 Born expansion of the Casimir–Polder interaction of a ground-state atom with dielectric bodies Appl. Phys. B 82 189–201
[45] Jackson J D 1975 Classical Electrodynamics (New York: Wiley)
[46] Andrews D L and Thirunamachandran T 1977 On three-dimensional rotational averages J. Chem. Phys. 67 3026–33