

Preprints are preliminary reports that have not undergone peer review. They should not be considered conclusive, used to inform clinical practice, or referenced by the media as validated information.

Mirror-mediated ultralong-range atomic dipoledipole interactions

Nicholas Furtak-Wells University of Leeds Benjamin Dawson University of Leeds Thomas Mann University of Leeds Gin Jose University of Leeds Almut Beige a.beige@leeds.ac.uk

University of Leeds

Research Article

Keywords:

Posted Date: April 4th, 2024

DOI: https://doi.org/10.21203/rs.3.rs-4193208/v1

License: (c) This work is licensed under a Creative Commons Attribution 4.0 International License. Read Full License

Additional Declarations: No competing interests reported.

Mirror-mediated ultralong-range atomic dipole-dipole interactions

Nicholas Furtak-Wells,¹ Benjamin Dawson,^{1,2} Thomas Mann,² Gin Jose,² and Almut Beige¹

¹ The School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom

²School of Chemical and Process Engineering, University of Leeds, Leeds LS2 9JT, United Kingdom

(Dated: March 30, 2024)

In three dimensions, dipole-dipole interactions which alter atomic level shifts and spontaneous decay rates only persist over distances comparable to the wavelength of the emitted light. To provide novel tools for quantum technology applications, like quantum sensing, many attempts have been made to extend the range of these interactions. In this paper we show that such an extension can be achieved with the help of partially transparent asymmetric mirror interfaces *without* involving negative refractive index metamaterials. Suppose two atoms are placed on opposite sides of the interface, each at the position of the mirror image of the other. In this case, their emitted light interferes exactly as it would when the atoms are right next to each other. Hence their dipole-dipole interaction assumes an additional maximum, even when the actual distance of the atoms is several orders of magnitude larger than their transition wavelength.

I. INTRODUCTION

In 1982, Scully and Drühl [1] proposed a double-slit experiment in which the slits are two two-level atoms. As illustrated in Fig. 1(a), the atoms are kept at a constant distance, are continuously driven by laser light and emit photons at a constant rate. When their distance is comparable to the wavelength of the emitted light, an interference pattern forms on a far-away screen. Averaged over many photons, this pattern very closely resembles the interference pattern of classical double-slit experiments. It only disappears when information about the origin of each photon becomes available [2]. As in classical two-slit interference experiments, the distance between the intensity minima and maxima depends on the distance between the atoms [3].

When this two-atom double-slit experiment was first performed by Eichmann *et al.* in 1993 [2], it raised many questions, like, how can spontaneously emitted photons interfere [4]. A closer look at the experiment shows that it is best *not* to think of the atoms as sources of spontaneously emitted photons but as continuously radiating dipole antennae [5]. Both atoms constantly transfer energy into the surrounding free radiation field which only manifests itself as "individual photons" upon detection [6, 7]. When an individual photon is registered on a photographic plate, it contains in general energy from both atoms. Depending on its direction of emission, radiation either interferes constructively or destructively, thereby resulting in a spatial dependence of the intensity of the emitted light. Moreover, interference effects result in a spatial dependence of first and second order photon correlations [8–10]. By now, the interference of light from distant atoms is relatively well understood and has already found applications in distributed quantum computing [11–15] and in quantum sensing [16].

Different from the classical case, interference in the two-atom double-slit experiment depends on the internal state of the slits, since different entangled atomic states radiate light in different preferred directions [17]. Suppose two atoms are right next to each other and share a single energy quantum. If the atoms in Fig. 1(a) are in their maximally-entangled symmetric state, all of the emitted light interferes constructively. The atomic coupling to the free radiation field is collectively enhanced and a photon is emitted at twice the usual rate. However, if the atoms are in their anti-symmetric state, their efforts to transfer their energy into the free radiation field cancel each other out. The spontaneous decay rate of the antisymmetric state therefore tends to zero. At larger distances between the atoms, similar alterations of spontaneous decay rates occur. These are synonymous with Dicke sub- and superrandiance [18] and indicate the presence of atomic dipole-dipole interactions [19–21]. A possible approach to atomic ultralong-range interactions is therefore the recreation of the interference effects of the original two-atom double-slit experiment for large atomic distances. Taking this into account, this paper derives a mirror-mediated, targeted ultralong-range atomic dipoledipole interaction which can persist over distances that are many orders of magnitude longer than the wavelength of the emitted light.

Suppose two atoms, a and b, are separated by a twosided partially transparent mirror and the position of each atom coincides with the position of the mirror image of the atom on the opposite side, as illustrated in Fig. 1(b). Comparing Figs. 1(a) and (b) and viewing the atoms again as radiating dipole antennae, we see that—for half of the emitted light—the two paths from a source to a certain point on the far-away screen are always of the same length. The resulting interference pattern is therefore the same as in the above described twoatom double-slit experiment [1], apart from a reduction in visibility. Since atomic dipole-dipole interactions are the result of interference effects and the interference of spontaneously emitted photons is the same in Figs. 1(a)and (b), the above discussion suggests an additional maximum of the dipole-dipole interaction between two atoms on opposite sides of a partially transparent mirror. As we shall see below, the strength of this ultralong-range interaction does not depend on the actual distance of the



FIG. 1. (a) Schematic view of the two-atom double-slit experiment by Eichmann et al. [2]. If the distance of the atoms is comparable to their transition wavelength λ_0 , the emitted light interferes either predominantly constructively or destructively. Which one applies depends on the collective state of the atoms. The result is an interference pattern on a far-away screen and a relatively short-range dipole-dipole interaction. As first pointed out by Dicke [18], the spontaneous decay rates of the atoms change (sub- and superradiance) and level shifts occur. (b) Schematic view of two atoms on opposite sides of a partially transparent asymmetric mirror which is smooth on one side but rough on the other. Such a mirror can be realised by placing tiny metallic islands onto a glass surface, while leaving small gaps between them. Now the transmitted light coming from atom a and the reflected light coming from atom b interfere exactly as in Fig. 1(a) and an analogous interference pattern emerges, if a screen is placed on the left hand side of the setup. However, the distance of the atoms no longer needs to be comparable to λ_0 for mirror-mediated dipole-dipole interactions to emerge. All that is required is that the distance between atom a and the mirror image of atom b is sufficiently small. (c) Unfortunately, constructive interference on the left hand side of the mirror usually implies destructive interference on the right and vice versa, thereby erasing any net effects of atomic dipole-dipole interactions. The spontaneous decay rates of collective atomic states therefore only changes, if the surface roughness of the mirror is not the same on both sides. For simplicity we consider in the following a partially transparent asymmetric metasurface which is smooth on one side but rough on the other such that there is effectively no interference between the reflected light coming from atom a and transmitted light coming from atom b.

atoms but on the distance between atom a and the mirror image of atom b.

Unfortunately, for symmetric mirrors, constructive interference of light on one-side implies destructive interference on the other [22]. Hence, as we shall see below, when interference effects due to the reflection of light on one side increase atomic interactions, reflection on the other side reduces them such that there is effectively no interaction. Remote dipole-dipole interactions therefore require the presence of an *asymmetric* mirror. To achieve this, we assume in the following that the mirror surface has a different surface roughness on both sides, as illustrated in Figs. 1(b) and (c). More concretely, we assume that the mirror surface is smooth on one side but uneven on the other. This can be realised, for example, by placing a thin layer of tiny metallic droplets onto a glass surface with some space between them. Although the atomic interactions which we predict here are weaker than the standard dipole-dipole interactions of nearby atoms, they are expected to find applications, for example, in non-invasive quantum sensing.

Atomic dipole-dipole interactions have already been studied in different environments but so far, they have always been relatively short-range [23, 24]. Theoretical and experimental studies usually consider atom-mirror interactions [25–28], interactions between atoms on the same side of an interface [29–31], atoms separated by negative index metamaterials and other thin films [32–40] and atoms near one-dimensional nanofibers and wave guides [41–44]. In this paper, we emphasise that, if a physical system can be modelled by classical physics, then the expectation values of the corresponding quantum model must evolve in exactly the same way. Otherwise, it would be possible to contradict quantum physics with classical experiments. Below we show that this observation can be used to simplify the derivation of quantum optical master equations, thereby enabling the modelling of the experimental setup shown in Fig. 1. All we need to know is, how classical optics models light propagation in the absence of the atoms but in the presence of the asymmetric mirror interface [45, 46].

II. RESULTS

A. Local atom-field interactions

In free space, the complex electric field observable $\mathcal{E}(\mathbf{r})$ at position \mathbf{r} can be written as a superposition of local contributions $\mathcal{E}_{s\lambda}(\mathbf{r})$ of travelling waves with polarisations $\lambda = 1, 2$ and directions of propagation \mathbf{s} ,

$$\boldsymbol{\mathcal{E}}(\boldsymbol{r}) = \sum_{\lambda=1,2} \int_{\mathcal{S}} \mathrm{d}\boldsymbol{s} \, \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}) \,. \tag{1}$$

Here S denotes the set of all three-dimensional unit vectors and $\mathcal{E}_{s\lambda}(\mathbf{r})$ creates local photons with wave vectors $\mathbf{k} = \mathbf{ks}$, normalised polarisation vectors $\mathbf{e}_{s\lambda}$ with $\mathbf{e}_{s1} \cdot \mathbf{e}_{s2} = \mathbf{e}_{s\lambda} \cdot \mathbf{s} = 0$, and bosonic creation operators $a_{k\lambda}^{\dagger}$. Using this notation, $\mathcal{E}_{s\lambda}(\mathbf{r})$ can be written as [48]

$$\boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}) = -\mathrm{i} \int_0^\infty \mathrm{d}k \, k^2 \, \left(\frac{\hbar c k}{16\pi^3 \varepsilon}\right)^{1/2} \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}} \, a_{\boldsymbol{k}\lambda}^\dagger \, \boldsymbol{e}_{\boldsymbol{s}\lambda} \, . \tag{2}$$

Suppose $|0_{\rm F}\rangle$ and $U_{\rm F}(t,0)$ denote the vacuum state and the time evolution operator of the free field Hamiltonian $H_{\rm F}$, respectively. Then

$$U_{\rm F}(t,0)\,\boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r})\,|0_{\rm F}\rangle = \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}+\boldsymbol{s}ct)\,|0_{\rm F}\rangle\,,\qquad(3)$$

since a local field excitation with a well defined direction of propagation s simply travels at the speed of light c in a straight line away from its source [45, 46]. If created at an initial time t = 0 at position r, it will be found at position r + sct at some later time t.

Next we assume that a partially transparent asymmetric metasurface is placed in the x = 0 plane, as illustrated in Fig. 1(b). Suppose this mirror is obtained by placing a thin metallic film with tiny holes which are much smaller than the wavelength of the emitted light onto a glass surface. In this case, the local field excitations which meet the gaps are transmitted and evolve exactly as they would in free space (i.e. as in Eq. (3)). However, light which does not meet a hole, is reflected and evolves such that

$$U_{\rm F}(t,0)\,\boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_b)\,|0_{\rm F}\rangle = \boldsymbol{\mathcal{E}}_{\tilde{\boldsymbol{s}}\lambda}(\tilde{\boldsymbol{r}}_b + \tilde{\boldsymbol{s}}ct)\,|0_{\rm F}\rangle\,,\qquad(4)$$

if is has been created at the position r_b of atom b at t = 0 and if the mirror surface is smooth on the left. The tilde indicates that a minus sign has been added to the x component of the respective vector, thereby ensuring for example that electric field vectors are always orthogonal to their direction of propagation. Similarly, if the metasurface is rough on the right, Eq. (3) changes into

$$U_{\rm F}(t,0)\,\boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_a)\,|0_{\rm F}\rangle = \boldsymbol{\mathcal{E}}_{\boldsymbol{S}(\boldsymbol{s})\lambda}(\boldsymbol{R}_a(\boldsymbol{s},t))\,|0_{\rm F}\rangle \qquad(5)$$

for reflected light originating from atom a at position r_a at t = 0. Here S(s) and $R_a(s,t)$ denote the direction of propagation and the position of the respective (s, λ) field excitation at time t. The exact values of these two variables does not need to be known, since we assume in the following that the surface roughness stops transmitted and reflected light from interfering efficiently on the right hand side of the mirror interface. The only assumption we make in the Methods section for simplicity is that the S(s) vectors cover the right hand side of the x = 0 plane relatively evenly.

In the following, we denote the electron charge and the complex dipole moment of atom *i* by *e* and $D_{12}^{(i)}$, respectively. Then, within the dipole and the rotating wave approximation, the interaction Hamiltonian between the atoms and the surrounding free radiation field can be written as [19–21]

$$H_{\rm AF} = e \sum_{i=a,b} \boldsymbol{D}_{12}^{(i)} \sigma_i^- \cdot \boldsymbol{\mathcal{E}}(\boldsymbol{r}_i) + \text{H.c.}$$
(6)

with σ_i^- denoting the atomic lowering operator. In Methods, we analyse the dynamics generated by this Hamiltonian using second order perturbation theory. As we shall see below, as long as we know how the atomic operators

 σ_i^- and the local electric field observable $\mathcal{E}_{s\lambda}(\mathbf{r}_i)$ evolve in the absence of atom-field interactions, the dynamics of the two atoms in Fig. 1 can be analysed in a relatively straightforward way.

B. Dynamics of atomic states

Quantum optical master equations describe the dynamics of atomic density matrices $\rho_{\rm A}(t)$ on a coarse grained time scale Δt which is much larger than their inverse transition frequency $1/\omega_0$ but also much smaller than their atomic lifetime $1/\Gamma_{\rm free}$ [6, 7]. To obtain them, we assume that the free radiation field is initially in its vacuum state $|0_{\rm F}\rangle$, evolve atoms and field for a time Δt with their Hamiltonian H in Eq. (11) and follow this dynamics with a measurement as to whether or not a photon has been emitted. Proceeding as described in Methods, one can then show that the time derivative of the atomic density matrix $\rho_{\rm A}$ equals

$$\dot{\rho}_{\rm A} = -\frac{\mathrm{i}}{\hbar} \left(H_{\rm cond} \rho_{\rm A} - \rho_{\rm A} H_{\rm cond}^{\dagger} \right) + \mathcal{L}(\rho_{\rm A}) \tag{7}$$

to a very good approximation. The reset operator $\mathcal{L}(\rho_{\rm A})$ and the non-Hermitian Hamiltonian $H_{\rm cond}$ in this equation can be used to analyse the dynamics of the two atoms in a time interval $(0, \Delta t)$ under the condition of a photon emission and no emission, respectively.

C. Dicke sub- and superradiance

Taking into account that some of the light that has been emitted by each atom travels to the opposite side of the mirror interface where it interferes with the reflected light originating from the atom on the opposite side, one can show that the operators $\mathcal{L}(\rho_{\rm A})$ and $H_{\rm cond}$ in Eq. (7) can be written as

$$\mathcal{L}(\rho_{\rm A}) = \sum_{i,j=a,b} \operatorname{Re}\left(\Gamma_{\rm mir}^{(ij)}\right) \sigma_i^- \rho_{\rm A} \sigma_j^+,$$

$$H_{\rm cond} = H_{\rm A} - \frac{\mathrm{i}\hbar}{2} \sum_{i,j=a,b} \Gamma_{\rm mir}^{(ij)} \sigma_j^+ \sigma_i^-. \tag{8}$$

The constants $\Gamma_{\min}^{(ij)}$ in these equations depend on the properties of the atoms and on the average reflection and transmission rates t_i and r_i of the mirror interface. In the following, we assume that these do not depend on the angle of incidence and the frequency of the incoming light. Such a dependence would alter the strength of the predicted interactions but we expect that our results remain valid, at least qualitatively.

Here we are especially interested in the case where the distance between atom a the mirror image of atom b, i.e. the difference between $\mathbf{r}_a = (x_a, y_a, z_a)$ and $\tilde{\mathbf{r}}_b = (-x_b, y_b, z_b)$, is very small. For simplicity, let us assume



FIG. 2. The imaginary part (a) and the real part (b) of $\Gamma_{\min}^{(ab)}$ in Eq. (9) as a function of the distance ξ between atom a and the mirror image of atom b for different orientations of the atomic dipole moment vectors $D_{12}^{(a)}$ and $D_{12}^{(b)}$ for $t_a r_b = 0.5$. For simplicity, we assume here that $D_{12}^{(a)} = D_{12}^{(b)} = D_{12}$ and that D_{12} is a real vector. The imaginary part is responsible for atomic level shifts of collective atomic states. Moreover, the real part of $\Gamma_{\min}^{(ab)}$ represents changes to the spontaneous decay rates in the presence of a mirror-mediated atomic dipole-dipole interaction.

that $y_a = y_b$ and $z_a = z_b$. In this case, the relative effective distance $\|\mathbf{r}_a - \tilde{\mathbf{r}}_b\|$ is simply given by $\xi = k_0(x_a + x_b)$. Using this notation and considering real mirror transmission and reflection rates for simplicity, one can show that

$$\Gamma_{\rm mir}^{(ab)} = \frac{3}{8} t_a r_b \Gamma_{\rm free} \left[\hat{\boldsymbol{D}}_{12}^{(a)} \cdot \hat{\boldsymbol{D}}_{12}^{(b)} \left(\frac{1}{i\xi} + \frac{1}{\xi^2} - \frac{1}{i\xi^3} \right) - \left(\hat{\boldsymbol{D}}_{12}^{(a)} \cdot \hat{\boldsymbol{x}} \right) \left(\hat{\boldsymbol{D}}_{12}^{(b)} \cdot \hat{\boldsymbol{x}} \right) \left(\frac{1}{i\xi} + \frac{3}{\xi^2} - \frac{3}{i\xi^3} \right) \right] e^{i\xi} - \frac{3}{16} t_a r_b \Gamma_{\rm free} \left[\hat{\boldsymbol{D}}_{12}^{(a)} \cdot \hat{\boldsymbol{D}}_{12}^{(b)} \left(\frac{1}{i\xi} - \frac{2}{i\xi^3} \right) + \left(\hat{\boldsymbol{D}}_{12}^{(a)} \cdot \hat{\boldsymbol{x}} \right) \left(\hat{\boldsymbol{D}}_{12}^{(b)} \cdot \hat{\boldsymbol{x}} \right) \left(\frac{1}{i\xi} + \frac{6}{i\xi^3} \right) \right], \quad (9)$$

while $\Gamma_{\text{mir}}^{(aa)} = \Gamma_{\text{mir}}^{(bb)} = \Gamma_{\text{free}}$ and $\Gamma_{\text{mir}}^{(ba)} = \Gamma_{\text{mir}}^{(ab)*}$. Here Γ_{free} denotes the single-atom free space decay rate and $\hat{D}_{12}^{(i)}$ and \hat{x} are unit vectors which point in the direction of the (real) dipole moment vector $D_{12}^{(i)}$ and of the positive x axis, respectively. Fig. 2 shows the imaginary and the real part of $\Gamma_{\text{mir}}^{(ab)}$ for different atomic dipole moments and for different distances ξ of the order of the wavelength of the emitted light. In the absence of a mirror interface, the reflection rate $r_b = 0$ and $\Gamma_{\text{mir}}^{(ab)}$ tends to zero, as one would expect. Formally, Eq. (7) is exactly the same as the master equations of two atoms experiencing Dicke sub- and superradiance [19–21]. The only difference is the overall factor $\frac{3}{8}t_ar_b$ in Eq. (9). In addition, there are some additional imaginary terms in the third and fourth line of this equation.

III. DISCUSSION

Having a closer look at the conditional Hamiltonian $H_{\rm cond}$ in Eq. (8), we see that it contains a Hermitian and a non-Hermitian contribution. The Hermitian contribution contains the atom Hamiltonian $H_{\rm A}$ and terms proportional to the imaginary part of $\Gamma_{\rm mir}^{(ab)}$. These describe the free dynamics of the atoms as well as interactioninduced level shifts. As one can see when comparing Eq. (9) with the equations in Refs. [18–21], the level shifts in the first two lines of Eq. (9) are essentially the same as the level shifts in the presence of free-space dipole-dipole interactions between two atoms at positions r_a and \tilde{r}_b . The only difference is the above mentioned overall factor, which occurs since not all emitted light contributes to the generation of the interaction. In addition there are some additional level shifts in the last two lines of Eq. (9). However, as illustrated in Fig. 2(a), these do not significantly alter the general dependence of the level shifts on the relative effective distance ξ of the two atoms. This is why we can refer to the interaction which we derive here as a dipole-dipole interaction.

The remaining terms in Eq. (8) describe the damping of population in excited atomic states. Diagonalising the conditional Hamiltonian $H_{\rm cond}$, we find that the spontaneous decay rate of the double-excited atomic state $|22\rangle$ equals $2\Gamma_{\rm free}$, as usual. However, collective atomic states which share only one excitation now have the spontaneous decay rates

$$\Gamma_{\pm} = \Gamma_{\rm free} \pm \operatorname{Re} \left(\Gamma_{\rm mir}^{(ab)} \right) \,. \tag{10}$$

As we can see from Eq. (9) and Fig. 2(b), up to an overall factor, the differences between Γ_{\pm} and Γ_{free} are what they would be in the presence of a free-space dipoledipole interaction between two atoms at positions r_a and \tilde{r}_b [18–21]. As shown in Section IV B, the atomic states with well-defined spontaneous decay rates are the same as for dipole-interacting atoms, namely the double-excited state |22 \rangle and the single-excited symmetric and antisymmetric states $|\pm\rangle$.

Changes to spontaneous decay rates can be detected, for example, with the help of fluorescence lifetime measurements. Moreover, when the atoms are driven by a common laser field, we expect their higher order photon correlation functions [8–10] to change and an interference pattern to emerge, if the spontaneously emitted photons are collected on a far-away screen, as illustrated in Fig. 1(b). As described in Methods, the only assumptions regarding the distance of the two atoms made in the derivation of the above equations are:

- 1. The actual distance between the atoms and between an atom and the mirror interface should be relatively large. This applies when $k_0|x_a - x_b| \gg 1$.
- 2. The actual distance between the two atoms in Figs. 1(b) and (c) should not be so large that the time it takes light to travel from one atom to the other becomes comparable to the lifetime of excited atomic states.

The first condition allows us to ignore direct atom-atom and atom-mirror interactions which are relatively shortrange. The second condition simplifies the modelling of light propagation in the presence of the mirror interface and is not very restrictive. For example, light can travel a 1 mm distance in less than $3.4 \cdot 10^{-12}$ s which is much shorter than the typical lifetime $1/\Gamma_{\text{free}}$ of excited atomic states. However, when analysing atomic interactions over very large distances, retardation effects need to be taken into account and the dynamics of the two atoms can no longer be described by a simple Markovian master equation, like the one in Eq. (7).

The main difference between the above-described mirror-mediated dipole-dipole interactions and the usual dipole-dipole interactions between two atoms in free space is that the former can be felt over much longer distances. As we have shown above, mirror-mediated remote dipole-dipole interaction can persist over distances which are several orders of magnitude longer than the wavelength of the emitted light. They assume a maximum when the relative distance ξ between the position \mathbf{r}_a of atom a and the position $\tilde{\mathbf{r}}_b$ of the mirror image of atom b is of the order of one. This can be the case even when the actual distance $||\mathbf{r}_a - \mathbf{r}_b||$ of the atoms is several orders of magnitude larger than the wavelength of the emitted light. The interaction which we predict here is therefore ultralong-range and targeted.

Another requirement for the atomic interactions which we predict in this paper is the presence of an *asymmetric* mirror interface. As illustrated in Fig. 1, this can be achieved with different surface roughness on both sides of the reflecting layer. If both sides of the mirror surface were smooth, the interaction constant $\Gamma_{\text{mir}}^{(ab)}$ in Eq. (9) would be proportional to $r_a t_b + t_a r_b$. Unfortunately, we know from classical optics (from Stoke's law which guarantees energy conservation) that this factor equals zero for standard partially transparent mirror interfaces [47]. For mirrors which are equally smooth on both sides, the interaction which we predict here therefore simply disappears. However, for the mirror interface shown in Figs. 1(b) and (c), constructive interference on one side of the mirror interface no longer implies the same amount of destructive interference on the other and the resulting interaction scales instead as $t_a r_b$.

In conclusion, in this paper we derived the quantum optical master equations of two atoms on opposite sides of a partially transparent asymmetric mirror interface by evolving the atoms and the free radiation field for a short time interval Δt using a Dyson series expansion. Our approach allows us to deduce the time derivative of the atomic density matrix ρ_A from the *classical* dynamics of light in the *absence* of any atom-field interactions. We then showed that the two atoms can experience an effective dipole-dipole interaction when atom a is placed close to the position of the mirror image of atom b and vice versa. The main result of this paper is the prediction of a targeted, remote, mirror-mediated ultralongrange dipole-dipole interaction which is likely to find a wide range of applications in the design of novel photonic devices for quantum technology applications, especially quantum sensing.

IV. METHODS

A. The derivation of quantum optical master equations

Our starting point for the derivation of the master equations in Eq. (7) is the relevant system Hamiltonian H which can be written as

$$H = H_{\rm A} + H_{\rm F} + H_{\rm AF} \,.$$
 (11)

Here $H_{\rm A}$ and $H_{\rm F}$ denote the free energy of the atoms and of the electromagnetic field, i.e. in the absence of the mirror interface. An expression for the interaction Hamiltonian $H_{\rm AF}$ between the atoms and the local excitations of the surrounding free radiation field within the usual dipole approximation can be found in Eq. (6) [19– 21]. As we shall see below, in addition, we only need to know how the atomic dipole moments and electric field observables evolve in the Heisenberg picture in the *absence* of atom-field interactions.

Suppose $\rho_{\rm A}(0)$ is the initial density matrix of the two atoms in the Schrödinger picture, while the surrounding free radiation field is initially in its vacuum state. We then evolve the atom-field density matrix $|0_{\rm F}\rangle\rho_{\rm A}(0)\langle 0_{\rm F}|$ for a time Δt with the time evolution operator $U(\Delta t, 0)$ of the above Hamiltonian H. Subsequently performing an absorbing measurement on the surrounding free radiation field leads to the atomic density matrix

$$\rho_{\rm A}(\Delta t) = \text{Tr}_{\rm F} \left[U(\Delta t, 0) | 0_{\rm F} \rangle \rho_{\rm A}(0) \langle 0_{\rm F} | U^{\dagger}(\Delta t, 0) \right].$$
(12)

Here the trace over the field degrees of freedom is taken to ensure that a measurement on the surrounding electromagnetic field does not change the properties of the atoms, if its outcome is ignored. As requested by locality, the density matrices $\rho_A(\Delta t)$ and $U(\Delta t, 0)|0_F\rangle\rho_A(0)\langle 0_F|U^{\dagger}(\Delta t, 0)$ must have the same atomic expectation values. Next, we introduce the time derivative

$$\dot{\rho}_{\rm A} = \frac{1}{\Delta t} \left(\rho_{\rm A}(\Delta t) - \rho_{\rm A}(0) \right) \tag{13}$$

which describes the dynamics of the atomic density $\rho_{\rm A}$ on the coarse grained time scale Δt under the condition that the free radiation field is essentially always kept in its vacuum state [6, 7].

Since the time evolution operator $U(\Delta t, 0)$ in Eq. (12) cannot be calculated easily analytically, we write the total Hamiltonian H of the experimental setup in Fig. 1(c) in the following as the sum of the free Hamiltonian $H_0 = H_A + H_F$ and the interaction H_{AF} . As long as Δt is neither too long nor too short, as described in Results, we can analyse the dynamics of the system using a Dyson series expansion which implies that

$$U(\Delta t, 0) = U_0(\Delta t, 0) - \frac{i}{\hbar} \int_0^{\Delta t} dt \, U_0(\Delta t, t) \, H_{AF} \, U_0(t, 0) - \frac{1}{\hbar^2} \int_0^{\Delta t} dt \int_0^t dt' \, U_0(\Delta t, t) \, H_{AF} \, U_0(t, t') \, H_{AF} \, U_0(t', 0)$$
(14)

to a very good approximation. Combining Eqs. (12) and (14), while only taking terms in zeroth order in Δt into account, leads to

$$\rho_{\rm A}(\Delta t) = \frac{1}{\hbar^2} \int_0^{\Delta t} dt \int_0^{\Delta t} dt' \operatorname{Tr}_{\rm F} \left[U_0(\Delta t, t) H_{\rm AF} U_0(t, 0) |0_{\rm F}\rangle \rho_{\rm A}(0) \langle 0_{\rm F}| U_0^{\dagger}(t', 0) H_{\rm AF} U_0^{\dagger}(\Delta t, t') \right]
- \frac{1}{\hbar^2} \int_0^{\Delta t} dt \int_0^t dt' \langle 0_{\rm F}| U_0(\Delta t, t) H_{\rm AF} U_0(t, t') H_{\rm AF} U_0(t', 0) |0_{\rm F}\rangle \rho_{\rm A}(0) + \text{c.c.}
+ \langle 0_{\rm F}| U_0(\Delta t, 0) |0_{\rm F}\rangle \rho_{\rm A}(0) \langle 0_{\rm F}| U_0^{\dagger}(\Delta t, 0) |0_{\rm F}\rangle \tag{15}$$

which applies in first order in Δt . To obtain the above equation, we took into account that $H_{\rm AF}$ either creates or annihilates a photon, while H_0 preserves the number of excitations in the free radiation field. Carefully comparing this equation with Eqs. (7) and (13), we see that

$$\mathcal{L}(\rho_{\rm A}) = \frac{1}{\hbar^2 \Delta t} \int_0^{\Delta t} dt \int_0^{\Delta t} dt' \operatorname{Tr}_{\rm F} \left[U_0(\Delta t, t) H_{\rm AF} U_0(t, 0) |0_{\rm F}\rangle \rho_{\rm A} \langle 0_{\rm F}| U_0^{\dagger}(t', 0) H_{\rm AF} U_0^{\dagger}(\Delta t, t') \right],$$

$$H_{\rm cond} = H_{\rm A} - \frac{\mathrm{i}}{\hbar \Delta t} \int_0^{\Delta t} dt \int_0^t dt' \langle 0_{\rm F}| U_0(\Delta t, t) H_{\rm AF} U_0(t, t') H_{\rm AF} U_0(t', 0) |0_{\rm F}\rangle.$$
(16)

To further simplify the above expressions, we notice that H_0 is the sum of two commuting Hamiltonians, namely H_A and H_F . Hence, $U_0(t,0) = U_A(t,0) \otimes U_F(t,0)$, where $U_A(t,0)$ and $U_F(t,0)$ denote the time evolution operators associated with H_A and H_F , respectively. In addition, we introduce the short hand notation

$$\boldsymbol{D}^{(i)}(t) = U_{\mathrm{A}}^{\dagger}(t,0) \, \boldsymbol{D}_{12}^{(i)} \sigma_i^- \, U_{\mathrm{A}}(t,0) \tag{17}$$

and notice that the vacuum state is invariant under $U_{\rm F}$. Hence, using Eqs. (6) and (16), one can show that

$$\mathcal{L}(\rho_{\mathrm{A}}) = \frac{e^{2}}{\hbar^{2}\Delta t} \sum_{i,j=a,b} \int_{0}^{\Delta t} \mathrm{d}t \int_{0}^{\Delta t} \mathrm{d}t' \operatorname{Tr}_{\mathrm{F}} \left[\boldsymbol{D}^{(i)}(t) \cdot U_{\mathrm{F}}(\Delta t, t) \boldsymbol{\mathcal{E}}(\boldsymbol{r}_{i}) |0_{\mathrm{F}}\rangle \rho_{\mathrm{A}} \langle 0_{\mathrm{F}} | \boldsymbol{D}^{(j)}(t')^{\dagger} \cdot \boldsymbol{\mathcal{E}}(\boldsymbol{r}_{j})^{\dagger} U_{\mathrm{F}}^{\dagger}(\Delta t, t') \right],$$

$$H_{\mathrm{cond}} = H_{\mathrm{A}} - \frac{\mathrm{i}e^{2}}{\hbar\Delta t} \sum_{i,j=a,b} \int_{0}^{\Delta t} \mathrm{d}t \int_{0}^{t} \mathrm{d}t' \boldsymbol{D}^{(j)}(t)^{\dagger} \cdot \langle 0_{\mathrm{F}} | \boldsymbol{\mathcal{E}}(\boldsymbol{r}_{j})^{\dagger} U_{\mathrm{F}}^{\dagger}(t', 0) \boldsymbol{D}^{(i)}(t') \cdot U_{\mathrm{F}}(t, 0) \boldsymbol{\mathcal{E}}(\boldsymbol{r}_{i}) |0_{\mathrm{F}}\rangle$$
(18)

in zeroth order in Δt . Here $\mathcal{L}(\rho_A)$ contains all the contributions of the atom-field density matrix which correspond to the presence of a photon at Δt in the free radiation field. It therefore equals the (unnormalised) density matrix of the atoms conditional on the creation of a photon in $(0, \Delta t)$. Analogously, the non-Hermitian Hamiltonian H_{cond} only contains contributions in which excitation has been created in $(0, \Delta t)$ but is later re-absorbed by the atoms. Hence it describes atomic dynamics in the absence of an emission [6, 7].

1. The free-space dynamics of atoms and field

Suppose $\hbar\omega_0$ is the energy gap between the ground and the excited state of atom *i*, i.e. between the states $|1\rangle_i$ and $|2\rangle_i$. In addition, we introduce the atomic raising and the lowering operators $\sigma_i^+ = |2\rangle_{ii}\langle 1|$ and $\sigma_i^- = |1\rangle_{ii}\langle 2|$ and write the atom Hamiltonian H_A as

$$H_{\rm A} = \sum_{i=a,b} \hbar \omega_0 \, \sigma_i^+ \sigma_i^- \,. \tag{19}$$

Hence the time-dependent dipole moment operator $\boldsymbol{D}^{(i)}(t)$ in Eq. (17) equals

$$\boldsymbol{D}^{(i)}(t) = e^{-i\omega_0 t} \, \boldsymbol{D}_{12}^{(i)} \, \sigma_i^- \,.$$
(20)

From Eq. (18) we see that the only other expression needed for the derivation of the quantum optical master equations in Eq. (7) is the state $U_{\rm F}(t, 0) \mathcal{E}_{s\lambda}(\mathbf{r}) |0_{\rm F}\rangle$ of the free radiation field. This state is obtained when creating a local field excitation with direction of propagation s and polarisation λ at time t = 0 at position \mathbf{r} and subsequently evolving the resulting state for some time t. Since Δt is much larger than the time it takes light to travel from the atoms to the mirror surface, light emitted at t = 0 in the direction of the mirror has either already been reflected or transmitted after almost all times $t \in (0, \Delta t)$. Neglecting very small times t for which light has not yet reached the mirror interface and using Eqs. (3) and (4), we therefore find that

$$U_{\rm F}(t,0) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_a) \left| 0_{\rm F} \right\rangle = \Theta(-s_x) \left[r_a(\boldsymbol{s}) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{S}(\boldsymbol{s})\lambda}(\boldsymbol{R}_a(\boldsymbol{s},t)) + t_a(\boldsymbol{s}) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_a + \boldsymbol{s}ct) \right] \left| 0_{\rm F} \right\rangle + \Theta(s_x) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_a + \boldsymbol{s}ct) \left| 0_{\rm F} \right\rangle, \\ U_{\rm F}(t,0) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_b) \left| 0_{\rm F} \right\rangle = \Theta(s_x) \left[r_b(\boldsymbol{s}) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{\tilde{s}}\lambda}(\tilde{\boldsymbol{r}}_b + \tilde{\boldsymbol{s}}ct) + t_b(\boldsymbol{s}) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_b + \boldsymbol{s}ct) \right] \left| 0_{\rm F} \right\rangle + \Theta(-s_x) \, \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_b + \boldsymbol{s}ct) \left| 0_{\rm F} \right\rangle$$
(21)

for direction vectors $\mathbf{s} = (s_x, s_y, s_z)$. Here the Heavyside function $\Theta(s_x)$ equals 0 for $s_x < 0$ and 1 otherwise. Moreover, the real reflection rates $r_i(\mathbf{s})$ for light travelling from atom i in the \mathbf{s} direction either equal 0 or 1, depending on whether light arrives at a metallic island or at a gap in the mirror interface (cf. Fig. 1). The corresponding transmission rates $t_i(\mathbf{s})$ are given by $t_i(\mathbf{s}) = 1 - r_i(\mathbf{s})$, since light with a well defined direction of propagation and source cannot be reflected and transmitted by the mirror surface. Later on, we will take into account that the effective reflection and transmission rates of the mirror are given by

$$r_i = \frac{1}{2\pi} \int_{\mathcal{S}_i} \mathrm{d}\boldsymbol{s} \, r_i(\boldsymbol{s}) \quad \text{and} \quad t_i = 1 - r_i \,, \tag{22}$$

where $S_a = \{s \in S : s_x < 0\}$ and $S_b = \{s \in S : s_x > 0\}$. As previously mentioned in the Results section, S(s) and $R_a(s,t)$ denote the direction of propagation and the position of a local electric field excitation at time t after its creation by atom a at t = 0 and after its subsequent reflection on the rough side of the mirror interface.

2. The conditional Hamiltonian H_{cond}

Substituting Eq. (20) into Eq. (18), we can show that the conditional Hamiltonian H_{cond} can indeed be written as in Eq. (8), if we define the constants $\Gamma_{\text{mir}}^{(ij)}$ such that

$$\Gamma_{\rm mir}^{(ij)} = \frac{1}{\Delta t} \int_0^{\Delta t} \mathrm{d}t \int_0^t \mathrm{d}t' \, \frac{e^2 c}{8\hbar\pi^3 \varepsilon} \,\mathrm{e}^{\mathrm{i}\omega_0(t-t')} \,\gamma_{\rm mir}^{(ij)}(t,t') \tag{23}$$

with $\gamma_{\min}^{(ij)}(t,t')$ given by

$$\gamma_{\rm mir}^{(ij)}(t,t') = \frac{16\pi^3\varepsilon}{\hbar c} \boldsymbol{D}_{12}^{(j)} \cdot \langle 0_{\rm F} | \boldsymbol{\mathcal{E}}(\boldsymbol{r}_j)^{\dagger} U_{\rm F}^{\dagger}(t',0) \boldsymbol{D}_{12}^{(i)} \cdot U_{\rm F}(t,0) \boldsymbol{\mathcal{E}}(\boldsymbol{r}_i) | 0_{\rm F} \rangle \,.$$
(24)

Using Eqs. (1), (2) and (21) and performing one of the k integrations, we therefore find that

$$\gamma_{\rm mir}^{(aa)}(t,t') = \sum_{\lambda=1,2} \int d\mathbf{k} \, k \, \mathrm{e}^{-\mathrm{i}ck(t-t')} \left[\left(t_a(\mathbf{s}) \,\Theta(-s_x) + \Theta(s_x) \right) \left(\mathbf{D}_{12}^{(a)} \cdot \mathbf{e}_{\mathbf{s}\lambda} \right)^2 + r_a(\mathbf{s}) \,\Theta(-s_x) \left(\mathbf{D}_{12}^{(a)} \cdot \mathbf{e}_{S(\mathbf{s})\lambda} \right)^2 \right] \\ \gamma_{\rm mir}^{(bb)}(t,t') = \sum_{\lambda=1,2} \int d\mathbf{k} \, k \, \mathrm{e}^{-\mathrm{i}ck(t-t')} \left[\left(\Theta(-s_x) + t_b(\mathbf{s}) \,\Theta(s_x) \right) \left(\mathbf{D}_{12}^{(b)} \cdot \mathbf{e}_{\mathbf{s}\lambda} \right)^2 + r_b(\mathbf{s}) \,\Theta(s_x) \left(\mathbf{D}_{12}^{(b)} \cdot \mathbf{e}_{\mathbf{\tilde{s}}\lambda} \right)^2 \right]$$
(25)

with $d\mathbf{k} = d\mathbf{s} dk k^2$. Next we introduce polar coordinates $k \in (0, \infty)$, $\varphi \in (0, 2\pi)$ and $\vartheta \in (0, \pi)$ such that

$$\boldsymbol{s} = \begin{pmatrix} \cos\vartheta\\ \cos\varphi\,\sin\vartheta\\ \sin\varphi\,\sin\vartheta \end{pmatrix}, \quad \boldsymbol{e}_{\boldsymbol{s}1} = \begin{pmatrix} 0\\ \sin\varphi\\ -\cos\varphi \end{pmatrix}, \quad \boldsymbol{e}_{\boldsymbol{s}2} = \begin{pmatrix} \sin\vartheta\\ -\cos\varphi\cos\vartheta\\ -\sin\varphi\cos\vartheta \end{pmatrix}, \quad (26)$$

while $ds = d\vartheta d\varphi \sin \vartheta$. After replacing the reflections and transmission rates $r_i(s)$ and $t_i(s)$ by their average values r_i and t_i in Eq. (22), which is well justified when the metallic islands which form the mirror interface are much smaller then the wavelength of the emitted light, Eq. (25) contains the integral

$$\int_{0}^{\infty} \mathrm{d}k \, k^{3} \, \mathrm{e}^{-\mathrm{i}ck\tau} = -\frac{\mathrm{i}\pi}{c^{4}} \, \delta^{(3)}(\tau) \tag{27}$$

with $\delta^{(3)}(\tau)$ denoting the third derivative of $\delta(\tau)$ with respect to τ . Hence we can now show that

$$\frac{1}{\Delta t} \int_0^{\Delta t} \mathrm{d}t \int_0^t \mathrm{d}t' \,\mathrm{e}^{\mathrm{i}\omega_0(t-t')} \int_0^\infty \mathrm{d}k \,k^3 \,\mathrm{e}^{-\mathrm{i}ck(t-t')} = \frac{\mathrm{i}\pi}{c^3 \Delta t} \int_0^{\Delta t} \mathrm{d}t \int_0^t \mathrm{d}\tau \,\mathrm{e}^{\mathrm{i}\omega_0\tau} \,\delta^{(3)}(\tau) = \frac{\pi \omega_0^3}{c^4} \,. \tag{28}$$

Combining the above equations and assuming that the direction vectors S(s) cover the half-space on the right hand side of the mirror interface evenly, we then find that

$$\Gamma_{\rm mir}^{(ii)} = \frac{e^2 \omega_0^3}{8\pi^2 \hbar \varepsilon c^3} \sum_{\lambda=1,2} \int d\boldsymbol{s} \left[r_i \,\Theta(\mp s_x) \left(\boldsymbol{D}_{12}^{(i)} \cdot \boldsymbol{e}_{\boldsymbol{\tilde{s}}\lambda} \right)^2 + \left(t_i \,\Theta(\mp s_x) + \Theta(\pm s_x) \right) \left(\boldsymbol{D}_{12}^{(i)} \cdot \boldsymbol{e}_{\boldsymbol{s}\lambda} \right)^2 \right],\tag{29}$$

respectively. Which signs apply depends on whether *i* equals *a* or *b*. Moreover, introducing the notation $\mathbf{D}_{12}^{(i)} = \|\mathbf{D}_{12}\| (d_1^{(i)}, d_2^{(i)}, d_3^{(i)})^{\mathrm{T}}$ with $|d_1^{(i)}|^2 + |d_2^{(i)}|^2 = 1$, one can now show that the above $\Gamma_{\min}^{(ii)}$ both equal the free-space decay rate of an atom with dipole moment $\mathbf{D}_{12}^{(i)} = \mathbf{D}_{12}$,

$$\Gamma_{\rm mir}^{(aa)} = \Gamma_{\rm mir}^{(bb)} = \Gamma_{\rm free} \quad \text{with} \quad \Gamma_{\rm free} = \frac{e^2 \omega_0^3 \|\boldsymbol{D}_{12}\|^2}{3\pi\hbar\varepsilon c^3} \tag{30}$$

since $r_i + t_i = 1$. As we shall see below, this result does not mean that photons are emitted at their free-space rate

 Γ_{free} , if initially only one of the two atoms is excited. The two remaining constants $\Gamma_{\text{mir}}^{(ab)}$ and $\Gamma_{\text{mir}}^{(ba)}$ in Eq. (8) can be derived analogously. Since we are only interested in the case where the distance of each atom from the mirror interface and the distance between atom a and atom b are much larger than the wavelength of the emitted light, we can safely ignore terms describing direct interactions between both atoms and between an atom and its own mirror image. These are known to be relatively short-range. However, terms describing interactions between an atom and the mirror image of the atom on the opposite side must be kept, when r_a and \tilde{r}_b are relatively close. Using Eqs. (1) and (24) we therefore find that

$$\gamma_{\min}^{(ab)}(t,t') = \frac{16\pi^{3}\varepsilon}{\hbar c} \sum_{\lambda=1,2} \int d\boldsymbol{s} \,\Theta(s_{x}) \bigg[t_{a}(\tilde{\boldsymbol{s}})r_{b}(\boldsymbol{s}) \,\boldsymbol{D}_{12}^{(b)} \cdot \langle 0_{\mathrm{F}} | \boldsymbol{\mathcal{E}}_{\tilde{\boldsymbol{s}}\lambda}(\tilde{\boldsymbol{r}}_{b} + \tilde{\boldsymbol{s}}ct')^{\dagger} \,\boldsymbol{D}_{12}^{(a)} \cdot \boldsymbol{\mathcal{E}}_{\tilde{\boldsymbol{s}}\lambda}(\boldsymbol{r}_{a} + \tilde{\boldsymbol{s}}ct) | 0_{\mathrm{F}} \rangle \\ + \int d\boldsymbol{s}' \,\Theta(-s'_{x}) \, r_{a}(\boldsymbol{s}')t_{b}(\boldsymbol{s}) \,\boldsymbol{D}_{12}^{(b)} \cdot \langle 0_{\mathrm{F}} | \boldsymbol{\mathcal{E}}_{\boldsymbol{s}\lambda}(\boldsymbol{r}_{b} + \boldsymbol{s}ct')^{\dagger} \,\boldsymbol{D}_{12}^{(a)} \cdot \boldsymbol{\mathcal{E}}_{\boldsymbol{S}(\boldsymbol{s}')\lambda}(\boldsymbol{R}_{a}(\boldsymbol{s}', t)) | 0_{\mathrm{F}} \rangle \bigg]$$
(31)

and $\gamma_{\min}^{(ba)}(t,t') = \gamma_{\min}^{(ab)}(t,t')^*$ to a very good approximation. If the scattering operator **S** scrambles the wave vectors of light originating from atom *a* more or less randomly upon reflection, the second term in this equation becomes negligible. However, the first term in the above equation does not average away and is in general non-zero. For simplicity, let us assume that r_a and r_b have the same y and z coordinates. In this case

$$e^{i\mathbf{k}\cdot(\mathbf{r}_a-\tilde{\mathbf{r}}_b)} = e^{ik\cos\vartheta(x_a+x_b)}.$$
(32)

Replacing $t_a(\tilde{s})$ and $r_b(s)$ by their average values in Eq. (22), proceeding as described in the previous subsection and using again Eq. (28), leads us to

$$\Gamma_{\rm mir}^{(ab)} = \frac{3t_a r_b \Gamma_{\rm free}}{8\pi} \sum_{\lambda=1,2} \int_{\pi/2}^{\pi} \mathrm{d}\vartheta \,\mathrm{e}^{-\mathrm{i}\xi\cos\vartheta} \,\sin\vartheta \int_{0}^{2\pi} \mathrm{d}\varphi \,\left(\hat{\boldsymbol{D}}_{12}^{(a)} \cdot \boldsymbol{e}_{\boldsymbol{s}\lambda}\right) \left(\hat{\boldsymbol{D}}_{12}^{(b)} \cdot \boldsymbol{e}_{\boldsymbol{s}\lambda}\right) \,. \tag{33}$$





FIG. 3. (a) Time dependence of the photon emission rate I(t) in Eq. (43) for different initial populations p of the excited atomic state in the presence of mirror-mediated dipole-dipole interactions. Here $\operatorname{Re}(\Gamma_{\min}^{(ab)}) = 0.05 \Gamma_{\text{free}}$. (b) The rate I(t) differs from the emission rate $I_0(t)$ of the atoms in the absence of interactions for the same p. As one would expect in the case of a broadening of spontaneous decay rates, the loss of atomic excitation happens faster at relatively short times and slower at later times.

The hat symbols indicate that the vectors $D_{12}^{(i)}$ have been normalised, the polarisation vectors $e_{s\lambda}$ can be found in Eq. (26), and $\xi = k_0(x_a + x_b)$ with $k_0 = \omega_0/c$ is a relative effective distance. Performing the φ integration and substituting $u = -\cos \vartheta$ yields

$$\Gamma_{\rm mir}^{(ab)} = \frac{3t_a r_b \Gamma_{\rm free}}{16} \int_0^1 \mathrm{d} u \,\mathrm{e}^{\mathrm{i}\xi u} \left[2d_1^{(a)} d_1^{(b)} \left(1 - u^2\right) + \left(d_2^{(a)} d_2^{(b)} + d_3^{(a)} d_3^{(b)}\right) \left(1 + u^2\right) \right]. \tag{34}$$

Performing the final integration, the above constant simplifies to

$$\Gamma_{\rm mir}^{(ab)} = \frac{3t_a r_b \Gamma_{\rm free}}{16} \left[\sum_{i=2}^3 d_i^{(a)} d_i^{(b)} \left(2e^{i\xi} \left(\frac{1}{i\xi} + \frac{1}{\xi^2} - \frac{1}{i\xi^3} \right) - \frac{1}{i\xi} + \frac{2}{i\xi^3} \right) - 2d_1^{(a)} d_1^{(b)} \left(2e^{i\xi} \left(\frac{1}{\xi^2} - \frac{1}{i\xi^3} \right) + \frac{1}{i\xi} + \frac{2}{i\xi^3} \right) \right]$$
(35)

which coincides with Eq. (9) in the main text. Analogously, one can show that $\Gamma_{\min}^{(ba)} = \Gamma_{\min}^{(ab)*}$. If the y and the z coordinates of the position of atom a and atom b are not the same, additional terms have to be taken into account in the above derivation. However, our physical intuition tells us that the remote interaction between atom a and atom b depends also in this case only on the relative effective distance ξ and not on the actual distance of the atoms.

3. The reset operator $\mathcal{L}(\rho_{\rm A})$

For completeness, we now also calculate the state $\mathcal{L}(\rho_A)$ of the atoms in case of an emission. Substituting Eq. (20) into Eq. (18) and introducing the variables $\tau = \Delta t - t$ and $\tau' = \Delta t - t'$ yields

$$\mathcal{L}(\rho_{\rm A}) = \sum_{i,j=a,b} \widetilde{\Gamma}_{\rm mir}^{(ij)} \sigma_i^- \rho_{\rm A} \sigma_j^+$$
(36)

with $\widetilde{\Gamma}_{\rm mir}^{(ij)}$ given by

$$\widetilde{\Gamma}_{\mathrm{mir}}^{(ij)} = \frac{e^2}{\hbar^2 \Delta t} \int_0^{\Delta t} \mathrm{d}\tau \int_0^{\Delta t} \mathrm{d}\tau' \,\mathrm{e}^{\mathrm{i}\omega_0(\tau-\tau')} \,\boldsymbol{D}_{12}^{(j)} \cdot \langle 0_{\mathrm{F}} | \boldsymbol{\mathcal{E}}(\boldsymbol{r}_j) \, U_{\mathrm{F}}^{\dagger}(\tau',0) \, \boldsymbol{D}_{12}^{(i)} \cdot U_{\mathrm{F}}(\tau,0) \boldsymbol{\mathcal{E}}(\boldsymbol{r}_i)^{\dagger} \, |0_{\mathrm{F}}\rangle \,. \tag{37}$$

These constants have many similarities with the constants $\Gamma_{\text{mir}}^{(ij)}$ in Eqs. (23). The only differences are a missing factor 2 and a different upper limit on the second time integral. Proceeding as in the previous subsection, we find that evaluating Eq. (37) now leads to time integrals of the form

$$\int_{0}^{\Delta t} \mathrm{d}\tau \int_{0}^{\Delta t} \mathrm{d}\tau' \,\mathrm{e}^{\mathrm{i}\omega(\tau-\tau')} = 2\mathrm{Re}\left(\int_{0}^{\Delta t} \mathrm{d}\tau \int_{0}^{\tau} \mathrm{d}\tau' \,\mathrm{e}^{\mathrm{i}\omega(\tau-\tau')}\right) \tag{38}$$

with $\omega = \omega_0 - ck$. Hence all the constants $\widetilde{\Gamma}_{\min}^{(ij)}$ are real and $\widetilde{\Gamma}_{\min}^{(ij)} = \operatorname{Re}\left(\Gamma_{\min}^{(ij)}\right)$ which yields the reset operator $\mathcal{L}(\rho_A)$ in Eq. (8).

B. Spontaneous decay of excited atomic states

To determine the spontaneous decay rates of two atoms on opposite sides of a partially transparent mirror interface, we denote their ground states by $|1\rangle$ and their excited states $|2\rangle$ and absorb all the Hermitian terms of the conditional Hamiltonian $H_{\rm cond}$ in Eq. (8) into the free atomic Hamiltonian $H_{\rm A}$. This is possible since these terms describe atomic level shifts. Doing so, the conditional Hamiltonian $H_{\rm cond}$ can be written as

$$H_{\text{cond}} = H_{\text{A}} - \frac{\mathrm{i}\hbar}{2} \left[\Gamma_{+} L_{+}^{\dagger} L_{+} + \Gamma_{-} L_{-}^{\dagger} L_{-} \right] \qquad (39)$$

where the Γ_{\pm} are the spontaneous decay rates of the two atoms in Eq. (10) and where the L_{\pm} with

$$L_{\pm} = (\sigma_a^- \pm \sigma_b^-) / \sqrt{2}$$
 (40)

are atomic lowering operators. Hence the time evolution operator $U_{\text{cond}}(t,0) = \exp(-iH_{\text{cond}}t/\hbar)$ which describes the dynamics of atom a and atom b under the condition of no photon emission in (0,t) equals, in the interaction picture with respect to $H_0 = H_A$ and t = 0,

$$U_{\text{cond}}(t,0) = |+\rangle \langle +| e^{-\Gamma_{+}t/2} + |-\rangle \langle -| e^{-\Gamma_{-}t/2} + |11\rangle \langle 11| + |22\rangle \langle 22| e^{-\Gamma_{\text{free}}t}.$$
(41)

Here the states $|\pm\rangle = (|12\rangle \pm |21\rangle/\sqrt{2}$ are the symmetric and the antisymmetric maximally entangled collective state of the atoms.

Suppose an incoherent excitation process prepares each atom with probability p in its excited state, thereby creating a statistical mixture of the atomic states $|11\rangle$, $|12\rangle$, $|21\rangle$ and $|22\rangle$. In this case, the probability $P_0(t) =$ $||U_{\text{cond}}(t,0)|\psi_1\rangle||^2$ for no photon emission in (0,t) is the sum of three exponentials and equals [6, 7]

$$P_{0}(t) = (1-p)^{2} + (1-p)p\left(e^{-\Gamma_{+}t} + e^{-\Gamma_{-}t}\right) + p^{2}e^{-2\Gamma_{\text{free}}t}.$$
(42)

For $p \ll 1$, the probability of finding both atoms in the excited state becomes negligible and the probability density I(t) for a photon emission at t coincides with the probability density $w_1(t) = -d/dt P_0(t)$ for the emission of a first photon at t. Hence,

$$I(t) = 2p \left[\Gamma_{\text{free}} \cosh \left(\text{Re} \left(\Gamma_{\text{mir}}^{(ab)} \right) t \right) - \text{Re} \left(\Gamma_{\text{mir}}^{(ab)} \right) \sinh \left(\text{Re} \left(\Gamma_{\text{mir}}^{(ab)} \right) t \right) \right] e^{-\Gamma_{\text{free}} t}$$
(43)

to a very good approximation. This equation holds up to first order in p. As illustrated in Fig. 3, this emission rate is qualitatively different from the emission rate $I_0(t)$ of the atoms in the absence of dipole-dipole interactions. It is therefore possible to use fluorescence lifetime measurements to detect the above described changes of spontaneous decay rates and to obtain a signature of the remote mirror-mediated dipole-dipole interactions which we predict in this paper.

Funding. We acknowledge financial support from the Oxford Quantum Technology Hub NQIT (grant number EP/M013243/1) and the EPSRC (2115757).

Competing Interest: The authors have no relevant financial or non-financial interests to disclose.

Author Contributions: Conceptualisation, G.J. and A.B.; methodology, N.F.-W., B.D. and A.B.; formal analysis, N.F.-W., B.D. and A.B.; writing and original draft preparation, N.F.-W., B.D. and A.B.; writing and review and editing, T. M. and G.J.; visualization, B.D. and A.B.; supervision, A.B.; funding acquisition, N.F.-W., G.J. and A.B. All authors have read and agreed to the published version of the manuscript.

Data availability: This publication is theoretical work that does not require supporting research data.

V. REFERENCES

- Scully, M. O. & Drühl, K. Quantum eraser: A proposed photon correlation experiment concerning observation and "delayed choice" in quantum mechanics. *Phys. Rev. A* 25, 2208 (1982).
- [2] Eichmann. U. et al. Young's interference experiment with light scattered from two atoms. *Phys. Rev. Lett.* **70**, 2359 (1993).
- [3] Schön, C. & Beige, A. Analysis of a two-atom double-

slit experiment based on environment-induced measurements. Phys. Rev. A **64**, 023806 (2001).

- [4] Beige, A., Schön, C. & Pachos, J. Interference of spontaneously emitted photons. *Fortschr. Phys.* 50, 594 (2002).
- [5] Beige, A. A new take on the slit experiment. Nat. Photon. 10, 290 (2016).
- [6] Hegerfeldt, G. C. How to reset an atom after a photon detection: Applications to photon counting processes.

Phys. Rev. A 47, 449 (1993).

- [7] Stokes, A. et al. Extending the validity range of quantum optical master equations. *Phys. Rev. A* 85, 053805 (2012).
- [8] Beige, A. & Hegerfeldt, G. C. Transition from antibunching to bunching for two dipole-interacting atoms. *Phys. Rev. A* 58, 4133 (1998).
- [9] Masson, S. J. et al. A. Many-Body Signatures of Collective Decay in Atomic Chains. *Phys. Rev. Lett.* 125, 263601 (2020).
- [10] Wolf, S. et al. Light of Two Atoms in Free Space: Bunching or Antibunching? *Phys. Rev. Lett.* **124**, 063603 (2020).
- [11] Lim, Y. L., Beige, A. & Kwek, L. C. Repeat-Until-Success Linear Optics Distributed Quantum Computing. *Phys. Rev. Lett.* **95**, 030505 (2005).
- [12] Barrett S. D. & Kok, P. Efficient high-fidelity quantum computation using matter qubits and linear optics. *Phys. Rev. A* **71**, 060310(R) (2005).
- [13] Duan, L.-M. & Monroe, C. Colloquium: Quantum networks with trapped ions. *Rev. Mod. Phys.* 82, 1209 (2010).
- [14] Hensen, B. et al. Loophole-free Bell inequality violation using electron spins separated by 1.3 kilometres. *Nature* 526, 682 (2015).
- [15] Stephenson, L. et al. High-rate high-fidelity entanglement of qubits across an elementary quantum network. *Phys. Rev. Lett.* **124**, 110501 (2020).
- [16] Lyons, A. et al. Fluorescence lifetime Hong-Ou-Mandel sensing. Nat. Comm. 14, 8005 (2023).
- [17] Araneda, G. et al. Interference of single photons emitted by entangled atoms in free space. *Phys. Rev. Lett.* **120**, 193603 (2018).
- [18] Dicke, R. H. Coherence in Spontaneous Radiation Processes. *Phys. Rev.* 93, 94 (1954).
- [19] Agarwal, G. S. Quantum statistical theories of spontaneous emission and their relation to other approaches. Springer Tracts in Modern Physics; Quantum Optics, Vol. **70** (Springer Verlag Berlin, 1974).
- [20] Gross, M. & Haroche, S. Superradiance: An essay on the theory of collective spontaneous emission. *Phys. Rep.* 93, 301 (1982).
- [21] Tanas R. & Ficek, Z. Entangled states and collective nonclassical effects in two-atom systems, *Phys. Rep.* **372**, 369 (2002).
- [22] Furtak-Wells, N. et al. Quantizing the electromagnetic field near two-sided semi-transparent mirrors. *Phys. Rev.* A 97, 043827 (2018).
- [23] Klimov, V. V. & Letokhov, V. S. Resonant energy exchange at nanoscale curved interface. *Chem. Phys. Lett.* 285, 313 (1998).
- [24] Klimov, V. V. & Letokhov, V. S. Resonant interaction between two atomic dipoles separated by the surface of a dielectric microsphere. *Phys. Rev. A* 58, 3235 (1998).
- [25] Drexhage, K. H. Influence of a dielectric interface on fluorescence decay time. J. Lum. 1, 693 (1970).
- [26] Worthing, P. T., Amos, R. M. & Barnes, W. L. Modification of the spontaneous emission rate of Eu³⁺ ions embedded within a dielectric layer above a silver mirror. *Phys. Rev. A* 59, 865 (1999).
- [27] Eschner, J. et al. Light interference from single atoms and their mirror images. *Nature* **413**, 495 (2001).

- [28] Hoi, I. C. et al. Probing the quantum vacuum with an artificial atom in front of a mirror. Nat. Phys. 11, 1045 (2015).
- [29] Palacino R. et al. Tuning the collective decay of two entangled emitters by means of a nearby surface. J. Phys. B 50, 1969 (2017).
- [30] Sinha, K., Venkatesh, B. P. & Meystre, P. Collective effects in Casimir-Polder forces. *Phys. Rev. Lett.* **121**, 183605 (2018).
- [31] Zhou, W., Rizzuto, L. & Passante, R. Vacuum fluctuations and radiation reaction contributions to the resonance dipole-dipole interaction between two atoms near a reflecting boundary. *Phys. Rev. A* 97, 042503 (2018).
- [32] Pendry, J. B. Negative Refraction Makes a Perfect Lens. Phys. Rev. Lett. 85, 3966 (2000).
- [33] Andrew, P. & Barnes, W. L. Förster Energy Transfer in an Optical Microcavity. *Science* 290, 785 (2000).
- [34] Andrew, P. & Barnes, W. L. Energy Transfer Across a Metal Film Mediated by Surface Plasmon Polaritons. *Sci*ence **306**, 1002 (2004).
- [35] Kästel, J. & Fleischauer, M. Suppression of spontaneous emission and superradiance over macroscopic distances in media with negative refraction. *Phys. Rev. A* 71, 011804(R) (2005).
- [36] Biehs, S.-A., Menon, V. M. & Agarwal, G. S. Long-range dipole-dipole interaction and anomalous Förster energy transfer across a hyperbolic metamaterial. *Phys. Rev. B*, 93, 245439 (2016).
- [37] Biehs, S.-A. & Agarwal, G. S. Qubit entanglement across ϵ -near-zero media. *Phys. Rev. A* **96**, 022308 (2017).
- [38] Deshmukh, R. et al. Long-Range Resonant Energy Transfer Using Optical Topological Transitions in metamaterials. ACS Photonics 5, 2737 (2018).
- [39] Newman, R. D. Observation of long-range dipole-dipole interactions in hyperbolic metamaterials. *Sci. Adv.* 4, eaar5278 (2018).
- [40] Karanikolos, V. Entanglement of quantum. emitters interacting through an ultra-thin noble metal nanodisk. *Opt. Express* 28, 24173 (2020).
- [41] Gonzalez-Tudela, A. et al. Entanglement of two qubits mediated by one-dimensional plasmonic waveguides. *Phys. Rev. Lett.* **106**, 020501 (2011).
- [42] Le Kien, F. & Rauschenbeutel, A. Propagation of nanofiber-guided light through an array of atoms. *Phys. Rev. A* **90** 063816 (2014).
- [43] Hung, C.-L. et al. Quantum spin dynamics with pairwisetunable, long-range interactions. *Proc. Natl. Acad. Sci.* U.S.A. 113, E4946 (2016).
- [44] Solano, P. et al. Super- and sub-radiance reveal infiniterange interactions through a nanofiber. Nat. Comm. 8, 1857 (2017).
- [45] Southall, J. et al. Locally-acting mirror Hamiltonians. J. Mod. Opt. 68, 647 (2021).
- [46] Hodgson, D. et al. Local photons. Front. Photon. 3, 978855 (2022).
- [47] Al Ghamdi, A. et al. A. Remote Non-Invasive Fabry-Perot Cavity Spectroscopy for Label-Free Sensing. *Sensors* 23, 385 (2023).
- [48] Bennett, R., Barlow T. M. & Beige, A. A physicallymotivated quantisation of the electromagnetic field. *Eur. J. Phys.* 37, 014001 (2016).