The independent scattering approximation in the saturated regime

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We show that a saturable single-frequency elastic T-matrix approach to scattering of light by atoms agrees remarkably well with a master equation description in the regime of unsaturated atoms, or for large separation between the atoms. If the atoms are in each other's near-field and the saturation of the atoms is high enough, the two approaches yield different results.

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I. INTRODUCTION

Applications of multiple scattering of light by point-scatterers are often based on the assumption that the scatterers under consideration scatter light independently from one another. All the scattering properties of each individual scatterer can then be expressed by a single mathematical object called the scatterer's T-matrix [1, 2]. This single-scatterer T-matrix is used as a building block in multiple-scattering theories [3] which can accurately describe many interesting phenomena [4, 5]. However, the use of T-matrices usually implies that one considers only elastic scattering. Although the restriction of scattering events to the pure elastic ones is justifiable in some cases, it is not necessarily correct for, e.g., scattering of high-intensity incident light fields [6].

In this paper, we will consider scattering of light by two atoms. The "atoms" could be implemented as any type of sub-wavelength quantum objects, for example: trapped atoms, quantum dots [7], trapped ions [8] or dye molecules. If we model the atoms as point-dipoles, we can use the T-matrix formalism to describe scattering of incident light. Alternatively, we can model the atoms as two-level systems, in which case multiple scattering of light can be studied using a master equation [9, 10, 11]. The system is then characterized by a density matrix, representing the coherences and populations of the atomic levels. Scattering of incident light then determines the evolution of the density matrix, while taking into account all orders of multiple scattering as well as inelastic scattering of light.

The goal of this paper is to compare the scattering properties of two atoms as described by a simple T-matrix approach and a (computationally much more involved) master equation approach. We will show that a saturable single-frequency elastic T-matrix approach agrees remarkably well with a master equation description in the regime of unsaturated atoms or for large separation between the atoms.

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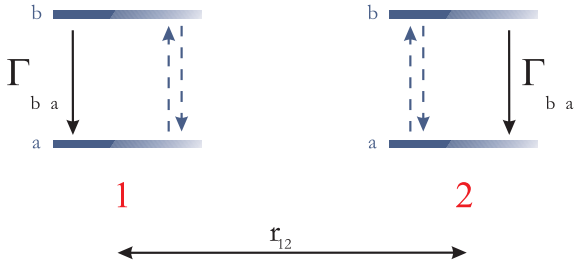


FIG. 1: The system under consideration: two identical atoms 1 and 2 are positioned in free space at a distance r_{12} from each other. Both atoms have a two-level internal structure. The upper level has a lifetime Γ_{ba}^{-1}. Both atoms interact with an incident field; this interaction is depicted by the dashed arrows.

The new aspect of this work is the comparison of two approaches which seem completely different at first but are in fact closely connected. We stress that the two approaches themselves, reviewed for clarity in sections II and III of our paper, have been extensively studied previously (see, e.g., [1, 9])

This paper is organized as follows. We start in Section II by describing the system we are considering. A master equation approach allows us to calculate how a near-resonant monochromatic incident field is scattered. In Section III, we consider the same system, and use a T-matrix approach to determine the scattering properties of the system. In Section IV we compare both methods. In Section V, finally, we discuss the applicability of our results to other atomic systems.

II. THE DENSITY MATRIX APPROACH TO SCATTERING OF LIGHT

We start by considering the system shown in Figure 1. Two identical atoms are positioned in free space, separated by a distance $r_{12} = |\boldsymbol{r}_{12}|$. Both atoms are modelled as a two-level system with upper level b and lower level a, separated by an energy difference $\hbar\omega_{ba}$. Decay between both levels occurs according to the decay

rate Γ_{ba}. A transition dipole moment $\boldsymbol{d}_{ab}^{i}$, $i \in \{1,2\}$ is associated with both $b \to a$ transitions. Both transition dipole moments are chosen to be equal in magnitude and orientation:

$$\boldsymbol{d}_{ab}^{i} \equiv d_{ab}\boldsymbol{\mu}, \qquad |\boldsymbol{\mu}| = 1, \qquad i \in \{1,2\}. \tag{1}$$

An incident monochromatic field $\boldsymbol{E}_0$ with wave vector $\boldsymbol{k}_0$ interacts with both atoms. The frequency $\omega_0 = |\boldsymbol{k}_0|c$ of the incident field is tunable and chosen near the $b \to a$ resonance (c is the velocity of light in free space). For simplicity, we have taken the wave vector $\boldsymbol{k}_0$ of the incident field perpendicular to $\boldsymbol{r}_{12}$. This choice of $\boldsymbol{k}_0$ ensures that both atoms always feel the same phase of the incident field, which makes the calculations somewhat easier without affecting the generality of our results.

We will now derive the master equation of the system; the following derivation closely follows [12]. The total Hamiltonian $\hat{H}_0$ describing the energies of the systems, the electromagnetic field and interactions is, in the standard electric dipole and rotating wave approximation, given by

$$\hat{H}_0 \equiv \hat{H}_D + \hat{H}_F + \hat{H}_{DL} + \hat{H}_{DF}. \tag{2}$$

The Hamiltonian of both individual atoms is given by

$$\hat{H}_D \equiv \hbar\omega_{ba}(\hat{S}_1^+\hat{S}_1^- + \hat{S}_2^+\hat{S}_2^-), \tag{3}$$

with $\hat{S}_i^{\pm}$ the dipole raising and lowering operators of atom i. Both atoms are coupled to the three-dimensional multimode electromagnetic field with Hamiltonian

$$\hat{H}_F \equiv \sum_{\boldsymbol{k}\lambda} \hbar\omega_{\boldsymbol{k}\lambda}\Big(\hat{a}_{\boldsymbol{k}\lambda}^{\dagger}\hat{a}_{\boldsymbol{k}\lambda} + \frac{1}{2}\Big), \tag{4}$$

where the operators $\hat{a}_{\boldsymbol{k}\lambda}^{\dagger}$ and $\hat{a}_{\boldsymbol{k}\lambda}$ respectively create and annihilate a photon in de mode $(\boldsymbol{k},\lambda)$. The modes of the electromagnetic field are taken to be in a vacuum state. Both atoms also interact through the Hamiltonian

$$\hat{H}_{DL} \equiv \frac{1}{2}\hbar\Omega\Big((\hat{S}_1^+ + \hat{S}_2^+)e^{i\omega_0 t} + \text{H.c.}\Big), \tag{5}$$

with an incident field $\boldsymbol{E}_0$ with frequency ω_0, where the Rabi frequency is given by $\Omega \equiv |\boldsymbol{d}_{ab}\cdot\boldsymbol{E}_0|/\hbar$. Finally, the atoms interact with the multimode vacuum field through the Hamiltonian

$$\hat{H}_{DF} \equiv \sum_{\boldsymbol{k}\lambda}\Big(\boldsymbol{\mu}\cdot(\boldsymbol{g}_{\boldsymbol{k}\lambda}(\boldsymbol{r}_1)\hat{S}_1^+ + \boldsymbol{g}_{\boldsymbol{k}\lambda}(\boldsymbol{r}_2)\hat{S}_2^+) + \text{H.c.}\Big), \tag{6}$$

where the mode function $\boldsymbol{g}_{\boldsymbol{k}\lambda}$ is given by

$$\boldsymbol{g}_{\boldsymbol{k}\lambda}(\boldsymbol{r}) \equiv \sqrt{\frac{\hbar\omega_{\boldsymbol{k}\lambda}}{2\varepsilon_0 L^3}}e^{i\boldsymbol{k}\cdot\boldsymbol{r}}\boldsymbol{\varepsilon}_{\boldsymbol{k}\lambda}, \tag{7}$$

with L^3 the quantization volume, ε_0 the vacuum permittivity and $\boldsymbol{\varepsilon}_{\boldsymbol{k}\lambda}$ the unit polarization vector of the field mode $(\boldsymbol{k},\lambda)$.

The dynamics of the total system can be expressed in terms of the Master equation for the density matrix $\hat{\sigma}$. The Master equation is, in the standard Born and Markov approximation, written in the Lindblad form, given by [12, 13]

$$\begin{aligned}\frac{d}{dt}\hat{\sigma} &= \hat{\mathcal{L}}\hat{\sigma} \\ &= \hat{\mathcal{L}}_{nd}\hat{\sigma} + \hat{\mathcal{L}}_{d}\hat{\sigma}.\end{aligned} \tag{8}$$

The non-dissipative part of the Lindblad operator can be written as

$$\hat{\mathcal{L}}_{nd}\hat{\sigma} \equiv -\frac{i}{\hbar}[\hat{H}_C + \hat{H}_0, \hat{\sigma}], \tag{9}$$

with coupling Hamiltonian

$$\hat{H}_C \equiv \hbar\delta^{(12)}\Big(\hat{S}_1^+\hat{S}_2^- + \hat{S}_2^+\hat{S}_1^-\Big), \tag{10}$$

while the dissipative part of the Master equation is given by

$$\begin{aligned}\hat{\mathcal{L}}_{d}\hat{\sigma} \equiv &-\frac{\Gamma_{ba}}{2}\Big(\hat{S}_1^+\hat{S}_1^-\hat{\sigma} - \hat{S}_1^-\hat{\sigma}\hat{S}_1^+ + \hat{S}_2^+\hat{S}_2^-\hat{\sigma} - \hat{S}_2^-\hat{\sigma}\hat{S}_2^+\Big) \\ &-\frac{\Gamma^{(12)}}{2}\Big(\hat{S}_1^+\hat{S}_2^-\hat{\sigma} + \hat{\sigma}\hat{S}_1^+\hat{S}_2^- - 2\hat{S}_2^-\hat{\sigma}\hat{S}_1^+\Big) + \text{H.c.}\end{aligned} \tag{11}$$

It is clear from equations (10) and (11) that the vacuum induces coupling between both atoms [14, 15, 16, 17]. The parameters $\delta^{(12)}$ and $\Gamma^{(12)}$ are not associated with individual systems, but with the total system as a whole. The cross-damping $\Gamma^{(12)}$ represents incoherent coupling between both atoms through spontaneous emission

$$\Gamma^{(12)} = \frac{2\pi}{\hbar^2}\sum_{\boldsymbol{k}\lambda}\Big(\boldsymbol{d}\cdot\boldsymbol{g}_{\boldsymbol{k}\lambda}(\boldsymbol{r}_1)\Big)\Big(\boldsymbol{d}\cdot\boldsymbol{g}_{\boldsymbol{k}\lambda}(\boldsymbol{r}_2)\Big)\delta(\omega_{ba}-\omega). \tag{12}$$

The parameter $\delta^{(12)}$ represents coherent coupling

through the vacuum, expressed by the frequency shift

$$\delta^{(12)} = -\frac{c}{4\pi}\mathcal{P}\int_{-\infty}^{+\infty}\frac{\omega^3}{\omega_{ba}^3}\Gamma^{(12)}\left(\frac{1}{\omega-\omega_{ba}}+\frac{1}{\omega+\omega_{ba}}\right)d\omega, \tag{13}$$

where $\mathcal{P}$ stands for Cauchy's Principal Value of the integral. Using standard evaluating techniques and replacing

$$\sum_{\mathbf{k}\lambda} \to \sum_{\lambda}\frac{L^3}{8\pi^3}\int_0^{+\infty}k^2dk\int_0^{2\pi}d\phi\int_0^{\pi}\sin(\theta)d\theta, \tag{14}$$

we find, in accordance [28] with [14]

$$\delta^{(12)} = \frac{3}{4}\Gamma_{ba}\Big(-\frac{1}{x}\cos(x)+\frac{1}{x^2}\sin(x)+\frac{1}{x^3}\cos(x)\Big),$$

$$\Gamma^{(12)} = \frac{3}{2}\Gamma_{ba}\Big(\frac{1}{x}\sin(x)+\frac{1}{x^2}\cos(x)-\frac{1}{x^3}\sin(x)\Big), \tag{15}$$

where we defined the dimensionless parameter $x \equiv \omega_{ba}r_{12}/c$, and where we used the fact that $\boldsymbol{\mu}\perp\boldsymbol{r}_{12}$. Both couplings $\delta^{(12)}$ and $\Gamma^{(12)}$ depend strongly on the distance between the atoms and the orientation of $\boldsymbol{\mu}$ and $\boldsymbol{r}_{12}$. In Figure 2 we show how $\delta^{(12)}$ and $\Gamma^{(12)}$ evolve as a function of x. For large separations ($x \gg 1$), the two couplings $\delta^{(12)}$ and $\Gamma^{(12)}$ vanish, independent of the orientation of the dipole moments. However, for small separations ($x \ll 1$), $\Gamma^{(12)}$ and $\delta^{(12)}$ exhibit a different behavior: while $\delta^{(12)}$ diverges for small separations, $\Gamma^{(12)}$ reduces to the single-atom decay rate Γ.

The standard method of solving the Master equation (8) is to calculate equations of motion for the density matrix elements and solve them by direct integration [18, 19]. This procedure means a set of 16 linear differential equations has to be solved. Using the resulting density matrix, we will calculate the total field, resulting from the scattering of $\boldsymbol{E}_0$. We therefore need to establish a relation between the total field and the calculated density matrix $\hat{\sigma}(t)$. Before establishing such connection, we note that from a didactic point of view, it is easier to connect both methods described in this paper if we calculate the total field in the far-field zone, i.e., $|\boldsymbol{r}| \gg r_{12}, (\omega_{ba}/c)^{-1}$. We stress, however, that our conclusions are also valid for observation points $\boldsymbol{r}$ in the near-field zone. The total electric field operator is given in the far-field by [10]

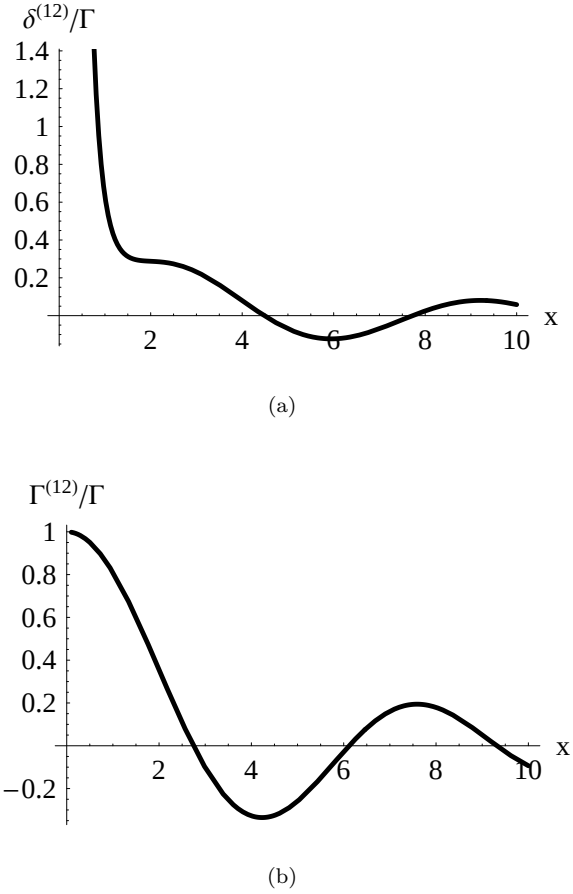


FIG. 2: The coherent coupling (Figure a) and the incoherent coupling (Figure b) (in units of Γ) as a function of the dimensionless parameter $x \equiv \omega_{ba}r_{12}/c$. We have chosen $\boldsymbol{\mu}$ perpendicular to $\boldsymbol{r}_{12}$, which is the case in the system studied in this paper.

$$\begin{aligned}\hat{\mathbf{E}}(\mathbf{r},t) &\equiv \hat{\mathbf{E}}^{+}(\mathbf{r},t)+\hat{\mathbf{E}}^{-}(\mathbf{r},t)\\ &= \left[\frac{1}{4\pi\varepsilon_0}\Big(\frac{\omega_{ba}}{c}\Big)^2\sum_{i=1,2}\frac{1}{|\mathbf{r}-\mathbf{r}_i|}(\mathbf{d}_{ab}-\mathbf{R}_i\frac{(\mathbf{R}_i\cdot\mathbf{d}_{ab})}{R_i^2})\times\right.\\ &\qquad\left.\hat{S}_i^{-}(t-|\mathbf{r}-\mathbf{r}_i|/c)\right]+\text{H.c.},\end{aligned} \tag{16}$$

where the shorthand notation $\mathbf{R}_i \equiv \mathbf{r}-\mathbf{r}_i$ has been used. Taking the expectation value and Fourier transforming yields

$$\left\langle \hat{\mathbf{E}}[\mathbf{r},\omega] \right\rangle = \int_{-\infty}^{+\infty} dt \left\langle \hat{\mathbf{E}}(\mathbf{r},t) \right\rangle e^{i\omega t}$$
$$= \Big(\frac{\omega_{ba}}{c}\Big)^2 \sum_{i=1,2} \frac{1}{|\mathbf{r}-\mathbf{r}_i|}(\mathbf{d}_{ab} - \mathbf{R}_i \frac{(\mathbf{R}_i \cdot \mathbf{d}_{ab})}{R_i^2}) \times$$
$$\frac{e^{i\frac{\omega}{c}|\mathbf{r}-\mathbf{r}_i|}}{4\pi\varepsilon_0} \int_{-\infty}^{+\infty} dt e^{i\omega t}\Big(\left\langle \hat{S}_i^-(t) \right\rangle + \text{H.c.}\Big). \tag{17}$$

If we now write (using the Green's funtion $\overleftrightarrow{G}_0$ defined as (23))

$$(\mathbf{d}_{ab} - \mathbf{R}_i \frac{(\mathbf{R}_i \cdot \mathbf{d}_{ab})}{R_i^2}) = -4\pi|\mathbf{r}| d_{ab} e^{-i\frac{\omega}{c}|\mathbf{r}|} \overleftrightarrow{G}_0(\mathbf{r}-\mathbf{r}_i,\omega)\cdot\boldsymbol{\mu}, \tag{18}$$

then

$$\left\langle \hat{\mathbf{E}}[\mathbf{r},\omega] \right\rangle = -\frac{d_{ab}}{\varepsilon_0}\Big(\frac{\omega_{ba}}{c}\Big)^2 \sum_{i=1,2} (\overleftrightarrow{G}_0(\mathbf{r}-\mathbf{r}_i,\omega)\cdot\boldsymbol{\mu}) \times$$
$$\int_{-\infty}^{+\infty} dt e^{i\omega t}\Big(\left\langle \hat{S}_i^-(t) \right\rangle + \text{H.c.}\Big). \tag{19}$$

In the steady-state regime, the coherences in the system (the off-diagonal elements of the density matrix) oscillate in phase with the incident field. We can easily study the behavior of the coherences by transforming to the interaction picture

$$\hat{\mathcal{S}}_i^+ \equiv e^{-i\omega_0 t}\hat{S}_i^+,$$
$$\hat{\mathcal{S}}_i^- \equiv e^{+i\omega_0 t}\hat{S}_i^-, \qquad i \in \{1,2\}, \tag{20}$$

such that the expectation values $\left\langle \hat{\mathcal{S}}_i^{\pm} \right\rangle$ are time-independent in the steady-state regime (see, e.g., [9]). The field component at $\omega = \omega_0$ (the frequency of the incident field) is then given by

$$\mathbf{E}[\mathbf{r},\omega_0] \equiv \left\langle \hat{\mathbf{E}}[\mathbf{r},\omega_0] \right\rangle$$
$$= -\frac{d_{ab}}{\varepsilon_0}\Big(\frac{\omega_{ba}}{c}\Big)^2 \sum_{i=1,2} (\overleftrightarrow{G}_0(\mathbf{r}-\mathbf{r}_i,\omega)\cdot\boldsymbol{\mu}) \left\langle \hat{\mathcal{S}}_i^- \right\rangle. \tag{21}$$

Expression (21) expresses the total field $\boldsymbol{E}$ on resonance as a function of the scattering properties of the atoms, without making any assumptions regarding the independence of both scatterers.

III. THE T-MATRIX APPROACH TO SCATTERING OF LIGHT

We now reconsider the system shown in Figure 1. We model both atoms as point-dipoles with dipole moments given by (1), and study the dynamics of the system using a T-matrix approach instead of a density matrix approach. The total field $\boldsymbol{E}$ at any position in the three-dimensional coordinate space can be written as the sum of the incident field and the field scattered by both atoms [20]:

$$\boldsymbol{E}[\omega,\boldsymbol{r}] = \boldsymbol{E}_0[\omega,\boldsymbol{r}]$$
$$+ \int\int \overleftrightarrow{G}_0(\omega,\boldsymbol{r}-\boldsymbol{s})\overleftrightarrow{T}(\omega,\boldsymbol{s},\boldsymbol{u})\boldsymbol{E}_0[\omega,\boldsymbol{u}]\boldsymbol{ds}\boldsymbol{du}, \tag{22}$$

where the integrations are over the full three-dimensional coordinate space. The 3×3 dyadic Green's function $\overleftrightarrow{G}_0(\omega,\boldsymbol{r})$ has as representation in coordinate space [21]:

$$\overleftrightarrow{G}_0(\omega,\boldsymbol{r})$$
$$= -(I^{(3)} + \frac{1}{\omega^2/c^2}\boldsymbol{\nabla}\otimes\boldsymbol{\nabla})\frac{e^{i\frac{\omega}{c}r}}{4\pi r}$$
$$= -\frac{e^{i\frac{\omega}{c}r}}{4\pi r}\Big(P(i\frac{\omega}{c}r)I^{(3)} + \tilde{P}(i\frac{\omega}{c}r)\boldsymbol{r}\otimes\boldsymbol{r}/r^2\Big), \tag{23}$$

with

$$P(z) = 1 - \frac{1}{z} + \frac{1}{z^2} \qquad \text{and} \qquad \tilde{P}(z) = -1 + \frac{3}{z} - \frac{3}{z^2}, \tag{24}$$

where $\otimes$ denotes the tensor product. The 3×3 unit tensor is denoted as $I^{(3)}$. We note that the Green's (23) function is related through (24) to the coherent coupling $\delta^{(12)}$ and incoherent coupling $\Gamma^{(12)}$ which arise in a master equation approach (see expressions (15)) by

$$\boldsymbol{\mu}\cdot\overleftrightarrow{G}_0(\omega,\boldsymbol{r}_1-\boldsymbol{r}_2)\cdot\boldsymbol{\mu} \equiv \frac{1}{6\pi}\frac{\omega_{ba}/c}{\Gamma_{ba}}\Big(2\delta^{(12)} - i\Gamma^{(12)}\Big), \tag{25}$$

indicating that coupling through the vacuum is intimately connected to ordinary free-space propagation. The T-matrix $\overleftrightarrow{T}$ of the total scattering system is given by

$$\overleftrightarrow{T}(\omega,\boldsymbol{r},\boldsymbol{r}') \equiv \langle\boldsymbol{r}|\overleftrightarrow{T}(\omega)|\boldsymbol{r}'\rangle$$
$$= \boldsymbol{\mu}\otimes\boldsymbol{\mu}\; t(\omega) \sum_{i,j=1,2} \delta(\boldsymbol{r}-\boldsymbol{r}_i)\delta(\boldsymbol{r}'-\boldsymbol{r}_j)\times$$
$$([I^{(2)} - \overleftrightarrow{D}(\omega,\boldsymbol{r}_1,\boldsymbol{r}_2)t(\omega)]^{-1})_{ij}, \tag{26}$$

where the delta-functions clearly depict the local character of both scatterers. The the 2×2 unit tensor is denoted by $I^{(2)}$. The 2×2 matrix $\overleftrightarrow{D}$ is given by

$$\overleftrightarrow{D}(\omega, \boldsymbol{r}_1, \boldsymbol{r}_2) \equiv \boldsymbol{\mu} \cdot \overleftrightarrow{G}_0(\omega, \boldsymbol{r}_1, \boldsymbol{r}_2) \cdot \boldsymbol{\mu} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{27}$$

Each atom has a T-matrix element $t(\omega)$ describing its scattering properties. In the system we are considering the T-matrix elements of both atoms are equal because the atoms are identical. In the limit of low-intensity incident fields, the T-matrix element $t(\omega)$ is given by the well-known expression for the dynamic polarizability of a two-level atom [22]

$$t(\omega) = \frac{3\pi}{\omega_{ba}/c} \frac{\Gamma_{ba}}{\left(\omega - \omega_{ba} + i\frac{\Gamma_{ba}}{2}\right)}. \tag{28}$$

For incident waves of higher intensities, both atoms will show saturation effects. The extension of expression (28) beyond the low-intensity limit is given by [23, 24]

$$t(\omega) = \frac{3\pi}{\omega_{ba}/c} \frac{\Gamma_{ba}}{\left(\omega - \omega_{ba} + i\frac{\Gamma_{ba}}{2}\right)} \frac{1}{1+s}, \tag{29}$$

which takes into account the loss of photons at frequency ω due to inelastic scattering; the inelastically scattered photons at frequencies $\omega' \neq \omega$ are not described. The saturation parameter s is defined as (see, e.g., [9])

$$s \equiv \frac{\Omega^2/2}{(\omega_{ba} - \omega)^2 + \Gamma^2/4}, \tag{30}$$

with the Rabi frequency $\Omega \equiv |\boldsymbol{d}_{ab} \cdot \boldsymbol{E}_0|/\hbar$. With the T-matrix element (28) we can rewrite the total field (22) as

$$\begin{aligned} \boldsymbol{E}[\omega, \boldsymbol{r}] =& \boldsymbol{E}_0[\omega, \boldsymbol{r}] + \frac{t(\omega)}{1 - t(\omega)\boldsymbol{\mu} \cdot \overleftrightarrow{G}_0(\omega, \boldsymbol{r}_1, \boldsymbol{r}_2) \cdot \boldsymbol{\mu}} \times \\ & \sum_{i,j=1,2} \overleftrightarrow{G}_0(\omega, \boldsymbol{r} - \boldsymbol{r}_i) \cdot \boldsymbol{\mu} \otimes \boldsymbol{\mu} \cdot \boldsymbol{E}_0[\omega, \boldsymbol{r}_j]. \end{aligned} \tag{31}$$

Equation (31) is the result of a T-matrix calculation, expressing the total field $\boldsymbol{E}$ as a function of the incident field $\boldsymbol{E}_0$ and the scattering properties of both atoms, explicitly assuming that they are independent from one another.

IV. COMPARISON OF BOTH METHODS AND DISCUSSION

We now compare expressions (21) and (31) and discuss their discrepancies. We consider three regimes.

A. Low-saturation incident field

If the saturation is very small ($s \ll 1$), the steady-state value of the density matrix can be calculated analytically. The resulting steady-state expectation value $\left\langle \hat{\mathcal{S}}_i^- \right\rangle$, $i \in \{1, 2\}$ is, using the linear expression (28),

$$\lim_{\Omega \to 0} \frac{1}{\Omega} \left\langle \hat{\mathcal{S}}_i^- \right\rangle = \frac{\omega_{ba}/c}{6\pi\Gamma_{ba}} \frac{t(\omega)}{1 - t(\omega)\boldsymbol{\mu} \cdot \overleftrightarrow{G}_0(\omega, \boldsymbol{r}_1, \boldsymbol{r}_2) \cdot \boldsymbol{\mu}}, \tag{32}$$

which yields, if substituted in (21), the same expression (31) as given by a T-matrix approach, hereby proving the validity of the T-matrix formalism for low-intensity incident fields for arbitrary separation between both atoms.

B. Atoms in each other's far-field: $r_{12} \gg (\omega_{ba}/c)^{-1}$

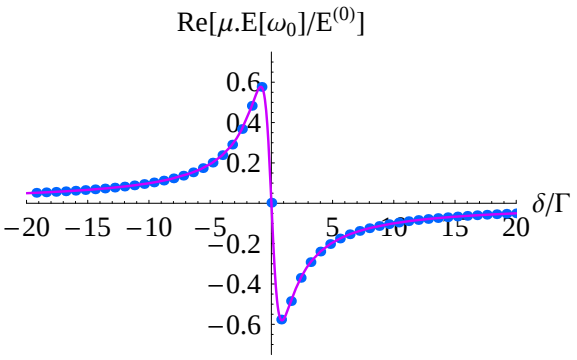
In the limit of large separation between both atoms $r_{12} \gg (\omega_{ba}/c)^{-1}$, both the coherent coupling $\delta^{(12)}$ and the incoherent coupling $\Gamma^{(12)}$ reduce to zero. In this regime, the density matrix $\hat{\sigma}$ obeys

$$\mathrm{Tr}_1(\hat{\sigma}) = \mathrm{Tr}_2(\hat{\sigma}) = \sigma_0, \tag{33}$$

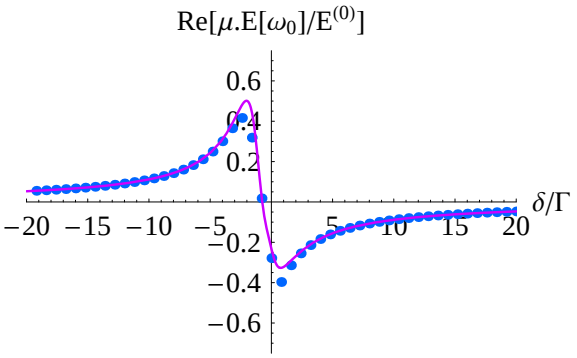
where σ_0 is the (2×2) density matrix corresponding to a single two-level system (see, e.g., [9]), and Tr_i stands for the trace over atom i. In other words, if both atoms are in each other's far-field, the single-atom density matrix factorizes out in two single-atom density matrices. This result corresponds with the intuitive expectation that the independent scattering approximation (31) is valid for large separation.

C. Arbitrary separation and incident field in the saturated regime

If the saturation parameter s is of order 1 or larger and the separation between the atoms is of the order of (or smaller than) $(\omega_{ba}/c)^{-1}$, no easy analytic expressions for the density matrix exist. However, the density matrix and the total field can still be evaluated numerically. In Figure 3 - 6, we have calculated $\boldsymbol{\mu} \cdot \boldsymbol{E}[\omega_0, \boldsymbol{r}]$ using expressions (21) and (31). Obviously, the numerical value of the total field component $\boldsymbol{\mu} \cdot \boldsymbol{E}[\omega_0, \boldsymbol{r}]$ depends on numerous parameters such as the incident field strength, the distance between both atoms, the evaluation point $\boldsymbol{r}$ and the frequency of the incident field. It is not our intention, however, to fully explore the entire $(\omega_{ba} - \omega_0, \boldsymbol{r}, r_{12}, \Omega)$ parameter space. We only want to point out that differences between both methods do arise, and explain why.

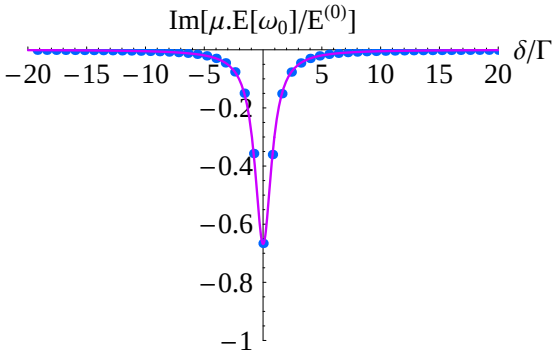


(a) $r_{12} = 100(\omega_{ba}/c)^{-1}$

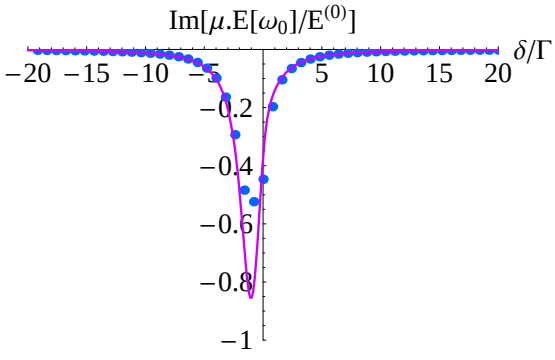


(b) $r_{12} = 0.8(\omega_{ba}/c)^{-1}$

FIG. 3: The real part of the field component $\boldsymbol{\mu}\cdot\boldsymbol{E}[\omega_0]/E^{(0)}$ on resonance, evaluated at the perpendicular bisector of $\boldsymbol{r}_{12}$. The Rabi frequency of the incident field is chosen $\Omega/\Gamma = 1$. The field has been calculated using both the T-matrix formalism (solid line) and the density matrix formalism (dotted line). The field has been plotted as a function of the detuning δ, for (a) atoms in each other's far-field, and for (b) atoms in each other's near-field.



(a) $r_{12} = 100(\omega_{ba}/c)^{-1}$



(b) $r_{12} = 0.8(\omega_{ba}/c)^{-1}$

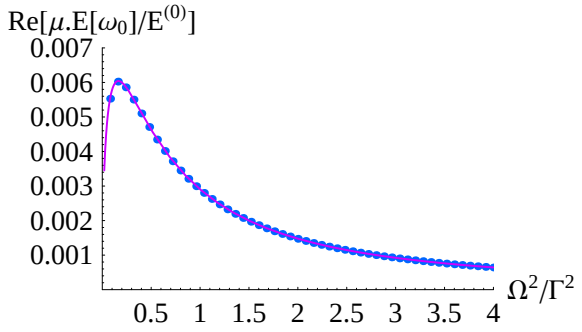
FIG. 4: The imaginary part of the field component $\boldsymbol{\mu}\cdot\boldsymbol{E}[\omega_0]/E^{(0)}$ on resonance, evaluated at the perpendicular bisector of $\boldsymbol{r}_{12}$. The Rabi frequency of the incident field is chosen $\Omega/\Gamma = 1$. The field has been calculated using both the T-matrix formalism (solid line) and the density matrix formalism (dotted line). The field has been plotted as a function of the detuning δ, for (a) atoms in each other's far-field, and for (b) atoms in each other's near-field.

As an example, the field has been evaluated at the perpendicular bisector of $\boldsymbol{r}_{12}$. The field is shown in units of

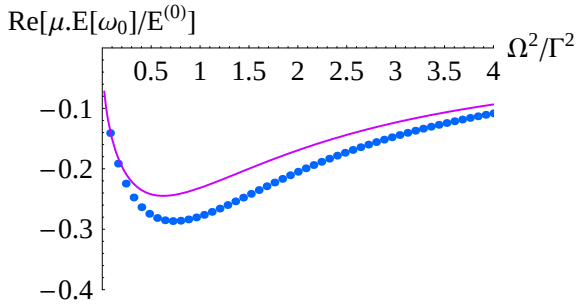
$$\boldsymbol{E}^{(0)} \equiv \frac{6\pi}{\omega_{ba}/c}\boldsymbol{\mu}\cdot\left(\overleftrightarrow{G}_0(\boldsymbol{r}-\boldsymbol{r}_1)+\overleftrightarrow{G}_0(\boldsymbol{r}-\boldsymbol{r}_2)\right)\cdot\boldsymbol{\mu}. \quad (34)$$

Figure 3 and 4 show the field as a function of the detuning $\delta \equiv 2(\omega_{ba}-\omega_0)$. Figure 5 and 6 show the field as a function of the Rabi frequency Ω, expressing the intensity of the incident field. It is clear that if both atoms are in each other's far-field ($r_{12} \gg (\omega_{ba}/c)^{-1}$), or if the saturation parameter is very small ($s \ll 1$), the predictions of the T-matrix approach and the density matrix approach coincide, as explained above. However, we see that for atoms in each other's near-field ($r_{12} \approx (\omega_{ba}/c)^{-1}$), the predictions of both approaches deviate for increasing saturation parameter of the incident field. At very high saturation ($s \gg 1$), both approaches again yield the same results.

We can explain the difference between both approaches as follows. For low-intensity incident fields, both atoms scatter light purely elastically. At higher intensities, more light is scattered inelastically (and less light elastically). In a T-matrix approach, the coupling of both atoms with inelastically scattered light is not taken into account. This explains why, in the case of a single scatterer, the T-matrix approach is exact, why for multiple scatterers, problems can arise when using T-matrices. The reason for this is that in the regime where the T-matrix approach breaks down, it is impossible to quantify the incident light on each scatterer, since the incident light to a certain extend consists of incoherent light, generated by multiple inelastic scattering between both

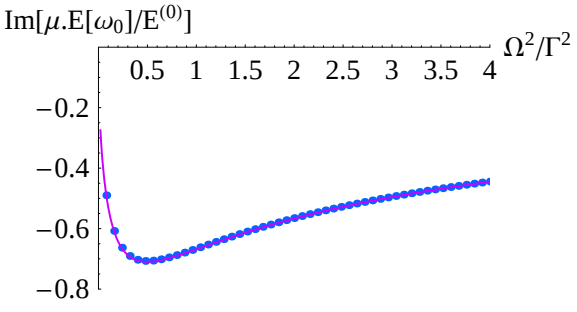


(a) $r_{12} = 100(\omega_{ba}/c)^{-1}$

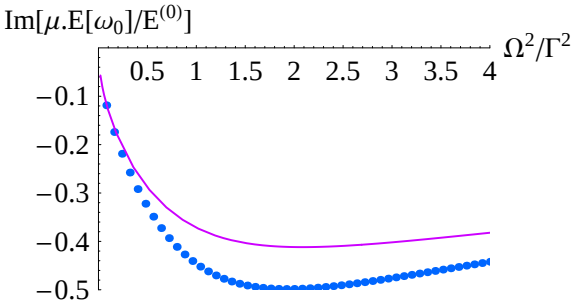


(b) $r_{12} = 0.8(\omega_{ba}/c)^{-1}$

FIG. 5: The real part of the field component $\boldsymbol{\mu} \cdot \boldsymbol{E}[\omega_0]/E^{(0)}$ on resonance, evaluated at the perpendicular bisector of $\boldsymbol{r}_{12}$. The detuning of the incident field is chosen $\delta = 0$. The field has been calculated using both the T-matrix formalism (solid line) and the density matrix formalism (dotted line). The field has been plotted as a function of the Rabi frequency Ω, for (a) atoms in each other's far-field, and for (b) atoms in each other's near-field.



(a) $r_{12} = 100(\omega_{ba}/c)^{-1}$



(b) $r_{12} = 0.8(\omega_{ba}/c)^{-1}$

FIG. 6: The imaginary part of the field component $\boldsymbol{\mu} \cdot \boldsymbol{E}[\omega_0]/E^{(0)}$ on resonance, evaluated at the perpendicular bisector of $\boldsymbol{r}_{12}$. The detuning of the incident field is chosen $\delta = 0$. The field has been calculated using both the T-matrix formalism (solid line) and the density matrix formalism (dotted line). The field has been plotted as a function of the Rabi frequency Ω, for (a) atoms in each other's far-field, and for (b) atoms in each other's near-field.

atoms. It is then no longer possible to define a single-scatterer scattering property, since coherences arise between different scatterers, which are induced by inelastically scattered light. Finally, at very high intensities of the incident field, both atoms will be completely saturated, regardless of the approach one uses to calculate the field. This saturation explains why for large s, the results produced by both methods again coincide.

V. EXTENSION

We stress that the breakdown of the independent scattering approximation we describe here applies to many other systems. In general, all systems where the contribution of incoherent light is not negligible will experience a breakdown of the independent scattering approximation. If, for example, one wishes to describe atomic systems with gain [23] or the effect of saturation on coherent backscattering [25, 26, 27] using independent atoms, one is limited to large separations between the atoms. If one wishes to describe these systems for atoms in each other's near-field, then the atoms cannot be described (from a light scattering point of view) as individual entities; they are to be considered as one large indivisible system. Only a description taking into account all self-induced internal coherences, such as a density-matrix description, then produces correct results.

VI. SUMMARY

In this paper, we studied the scattering properties of a system consisting of two atoms in free space. The scattering of incident monochromatic light has been considered, using both a saturable single-frequency elastic T-matrix approach and a density matrix approach. The former method assumes both atoms to be independent, while the latter method does not make such initial assumption. We found that in the regime of low saturation parameters, or for large distance between both atoms, both approaches yield the same predictions. However, if the atoms are in each other's near-field and the saturation parameter is high enough, the two approaches produce different results. This discrepancy is due inelastic scattering of light, which is not taken into account in a T-matrix approach.

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[28] note a small printing error in [14] in the expression for $\Gamma^{(12)}$