

Classically tuning the quantum interference of two photons scattered by a macroscopic lossy sphere

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We investigate the quantum optical scattering of two-photon wave packets by a macroscopic lossy sphere by means of macroscopic quantum electrodynamics in the form of modified Langevin noise formalism. Building on our recently developed general scattering framework, we show that under the far-field approximation, the probability amplitudes for photon detection exhibit a direct correspondence to the classical electric field. This allows us to effectively map the quantum interference calculation for the spherical geometry onto a solvable classical electromagnetic problem, specifically by expressing the quantum-scattering amplitudes in terms of the classical Mie coefficients. The two ingoing photons with an arbitrary frequency-polarization spectrum impinge onto the sphere along two different directions and, as a consequence of matter losses, their scattering involves the three independent processes where two, one, and zero outgoing photons survive. Noncollinearity of ingoing photons causes the existence of two different quantum paths they can follow upon scattering, producing interference effects in the detection of the above three processes, which is governed by the wave-packet spectral symmetry. Furthermore, we demonstrate that the sphere's rotational and reflectional symmetries can be exploited to identify specific scattering geometries that enforce perfect constructive or destructive (Hong-Ou-Mandel) interference, both for symmetric and antisymmetric wave packets. To assess the impact of matter dispersion or losses on quantum interference effects accompanying photon detection, we analyze the scattering of narrow-band two-photon wave packets by high-index dielectric lossy spheres. We show that classical Mie resonance peaks, due to their Fano-like traits, yield very strong constructive and destructive interference effects, occurring when the wave packet carrier frequency matches the resonance frequency and side Fano dip frequency, respectively. In addition, we consider the overall scattering probabilities of two, one, and zero photons, and we prove that, at the Mie resonance frequencies, they exhibit quantum interference effects that are extremely sensitive to the spectral symmetry of the input wave packet, thus suggesting an efficient spectral technique assisted by matter losses to identify entanglement.

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I. INTRODUCTION

Multiphoton interferometry [1] is among the most relevant research topics in quantum optics, both for its conceptual interest stemming from the intimate connection with photon indistinguishability, and for its far-reaching applications in quantum computation [2] which hinge upon the marked sensitivity to entanglement. The interfering quantum paths are usually provided in literature by lossless optical devices as beam splitters [3–6], general passive optical systems [7] and planar devices [8–11] which essentially couple their input and output photonic modes without altering the energy balance, i.e., they are characterized by a unitary relation joining the boson operators of output modes to those of the input ones. Lossless beam splitters are able to perform two-photon interference [12–19] which is probably the most paradigmatic quantum interferometric phenomenon, encompassing

the limiting effects of photon coalescence, or Hong-Ou-Mandel (HOM) effect, and photon anticoalescence resulting from input two-photon wave packets whose spectrum is symmetric and antisymmetric, respectively. On the other hand, other lossless devices, such as Mach-Zehnder interferometers [20,21], plasmonic circuits [22–24], silicon chips [25] and q plates [26,27] have been used to produce two-photon interference, all mimicking the archetypal functionality of lossless beam splitters. In addition, lossless devices with more than two input and output ports [28–31] have opened the possibility to observe multiphoton interference. When losses cannot be neglected, optical devices still couple their input and output photonic modes but radiation-matter energy exchange requires to account for matter modes as well, so that the device is characterized by a unitary input-output relation involving both photon and matter (Langevin) boson operators [32,33], as specifically reported for four-port [34] and N -port [35] devices, multilayer dielectric plates [36] and gratings [37]. The simplest but fundamental setup of a lossy beam splitter fed by a two-photon wave packet has been investigated in Refs. [38,39] where the authors proved that conditions still exist to observe the HOM effect. In the same paper, the authors also showed that, as a consequence of matter losses, the beam splitter functionality involves the additional processes where one or zero photons survives and that, remarkably, a

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further quantum interference effect (apparent nonlinear absorption) exists, which is able to suppress the one-photon survival process. However, in Ref. [40] the authors argued theoretically that the phenomenology of two-photon interference produced by lossy devices is much more rich since the additional freedom provided by losses in designing the device can be harnessed to engineer the interference, making it range from photon coalescence to anticoalescence regardless of the spectral symmetry of the input wave packet. This is possible since losses literally create otherwise-not-existing quantum paths, as experimentally confirmed in various setups [41–46].

Theoretical predictions concerning two-photon interference rely on the quantum description of the optical device functionality, in turn obtained from a theoretical scheme accounting for the interaction of the quantized electromagnetic field with macroscopic matter. While macroscopic classical electrodynamics is rather simply quantized when only transparent dielectrics are involved [47,48], it is considerably harder to turn it into a quantum theory when optical absorption cannot be neglected, as a consequence of the necessary inclusion in the theory of the matter degrees of freedom [49–54]. Langevin noise formalism (LNF) [55–59], or macroscopic quantum electrodynamics (MQED), is nowadays the most general theoretical framework for modeling the quantum optical field inside a spatially unbounded absorbing medium, as proved by its many and different applications [60–70]. Basically, LNF regards the field as entirely due to dipolar medium sources whose quanta, the electric and magnetic bosonic polaritons, provide the quantum field description. On the other hand, if the absorbing medium is confined within a finite-size object, LNF can still be used by resorting to a singular limiting procedure consisting in taking the permittivity and permeability limits $\text{Im}(\epsilon) \rightarrow 0^+$ and $\text{Im}(\mu) \rightarrow 0^+$ pertaining the regions filled by vacuum at the end of the calculations [71]. Recently, an alternative version of MQED has been introduced, the modified Langevin noise formalism (MLNF) [72,73], which avoids the singular limiting procedure when dealing with lossy objects of finite size in vacuum. This is possible since, in the MLNF, the field displays both the contributions of the dipolar medium sources and of the scattering modes, so that the quantum portrayal is provided by electric (e), magnetic (m), and scattering (s) bosonic polaritons, the scattering polaritons being the counterparts of standard photons in the lossy regime. Note that, in the lossless limit, the MLNF coincides with Glauber’s approach to quantum optics of dielectric media of Ref. [47] since e and m polaritons disappear and s polaritons reduce to standard photons. The MLNF flexibility to deal with the quantized scattering modes has recently been exploited to model the interaction of quantum emitters with dispersive objects [74] and to develop a general approach to quantum optical scattering by finite lossy objects in vacuum [75].

In this paper, we investigate various quantum interference effects accompanying the scattering of two-photon wave packets by a nonmagnetic macroscopic lossy sphere by resorting to the general approach of Ref. [75]. While our previous work established the general theoretical framework, here we employ specific approximations—namely, the

far-field and narrow-bandwidth limits—to reduce the modified Langevin noise formalism into a computationally tractable framework. This derivation connects the first-principle theory to a practical calculation scheme, allowing quantum interference observables for the spherical geometry to be analytically evaluated using classical Mie scattering coefficients. It is worth pointing out that the only quantum optical theoretical analysis dealing with a macroscopic sphere is, to the best of our knowledge, the one reported in Ref. [76] where the sphere is assumed to be lossless and dispersionless, as opposed to the present paper where we mainly focus on the role played by matter losses on quantum interference effects. After reviewing the general formalism (Sec. II), we consider the scattering by an arbitrary object of two ingoing s polaritons (the photon counterparts in the presence of lossy matter, in MLNF) traveling along different directions and with an arbitrary frequency-polarization wave-packet spectrum (Sec. III). Since matter absorption can destroy an ingoing s polariton with the creation of an e polariton (i.e., a quantized medium dipolar excitation), the outgoing quantum radiation state involves three contributions, respectively containing two s , one s and one e , and two e outgoing polaritons. The corresponding ss , se , and ee processes closely parallel those supported by a lossy beam splitter and discussed in Refs. [38,39] with the essential difference that, in our case, the directions of the two ingoing s polaritons and those of the detected outgoing s polaritons can be arbitrarily chosen, so that the scatterer object can effectively be regarded as an optical device with an infinite number of ports. The quantum amplitudes of the three processes turn out to be the superposition of a direct and an exchange term, which correspond to the two possible quantum paths connecting the ingoing polaritons to the outgoing ones. Accordingly the ss , se , and ee processes display interference effects that are maximal for an ingoing wave packet with symmetric or antisymmetric spectra, so that we here focus on these two relevant cases by analyzing the ensuing probability density of polariton-coincidence detection and overall scattering probabilities of two, one, and zero polaritons. After specializing the treatment to a macroscopic lossy sphere (Sec. IV), we exploit the ensuing full rotational invariance to discuss two classes of scattering geometries such that the polariton-coincidence detection exhibits perfect constructive or destructive interference, depending on the wave-packet symmetry (the former effect substantially representing a far-reaching generalization of the famous Hong-Ou-Mandel effect in 50-50 beam splitters; see Sec. V). To discuss the role played by matter dispersion and losses of quantum interference effects, we focus on subwavelength lossy spheres with high-index dielectric behavior and, using narrow-band ingoing wave packets, we analyze the spectral features produced by such spheres in the polaritons detection (Sec. VI). As a main general result, we prove that the classic Mie resonances of the sphere support a very efficient quantum interference mechanism accompanying any measurement concerning the scattered radiation. In particular, in the probability density of polariton-coincidence detection, we find that the well-known Fano-like spectral profile of the Mie resonance peaks [77,78] is responsible for very marked constructive or destructive interference, respectively occurring when the wave-packet

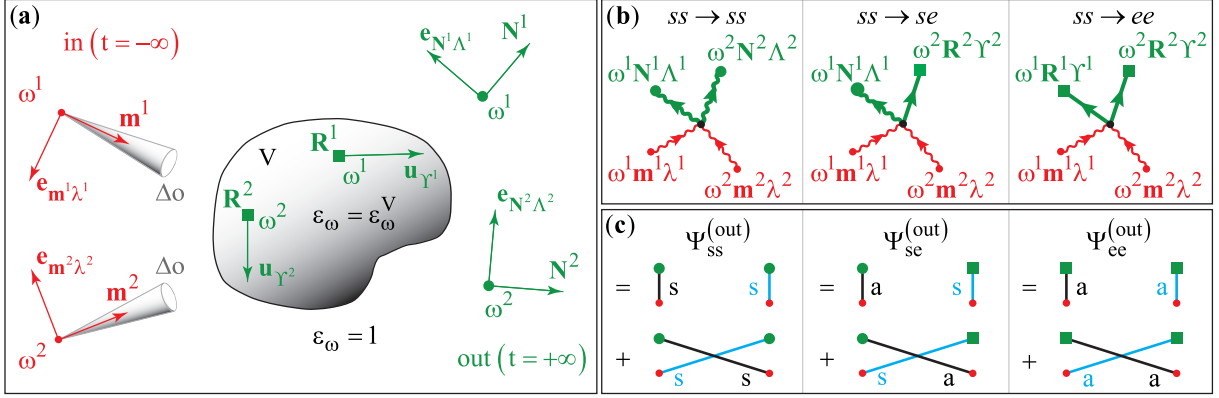


FIG. 1. Schematic representation of the quantum optical scattering of a two-photon wave packet by a macroscopic lossy object. (a) General geometric setup: a target object of volume V and permittivity ϵ_ω is placed in vacuum. In the far past ($t = -\infty$), the input state consists of two incident s polaritons (represented by red dots) filling the small solid angles $\Delta\omega$ around the central directions $\mathbf{m}^1$ and $\mathbf{m}^2$. In the far future ($t = +\infty$), the scattered field is characterized by outgoing s polaritons (larger green dots) and outgoing e polaritons (green squares), detected along directions $\mathbf{N}^1$ and $\mathbf{N}^2$ with specific polarizations $\mathbf{e}_{\mathbf{N}^1\Lambda^1}$ and $\mathbf{e}_{\mathbf{N}^2\Lambda^2}$. Note that the same color and shape coding for the polaritons (red dots for ingoing s , green dots for outgoing s , and green squares for outgoing e) is consistently used in panels (b) and (c). (b) Schematic of the three global scattering channels: two-polariton survival ($ss \rightarrow ss$), single-polariton survival ($ss \rightarrow se$), and complete absorption ($ss \rightarrow ee$). (c) Graphical representation of the three probability amplitudes, namely, $\Psi_{ss}^{(\text{out})}$, $\Psi_{se}^{(\text{out})}$, and $\Psi_{ee}^{(\text{out})}$, corresponding to the scattering processes depicted in panel (b) [and described by Eq. (8)]. Each subpanel in panel (c) corresponds to the scattering channel depicted directly above it in panel (b). In panel (c), for each amplitude, the first row (after the = sign) represents the direct term in Eq. (8), while the second row (after the + sign) represents the exchange term. The labels s and a denote the corresponding coefficients appearing in the analytical expressions of Eq. (8).

central frequency nearly matches the resonance frequency or the side Fano dip frequency. Besides, in the overall scattering probabilities of two, one, and zero s -polaritons, we find that the wave-packet symmetry plays a dominant role at isolated Mie resonances where the scattering probabilities for symmetric and antisymmetric wave packets can differ by several orders of magnitude. This suggests that classical resonances can provide efficient alternative ways to detect polariton entanglement. Finally, we discuss the experimental feasibility and the required conditions for observing these quantum interference effects (Sec. VII), before summarizing our main findings in the concluding remarks (Sec. VIII).

II. MAIN FORMALISM

We start by reviewing the MLNF detailed in Ref. [73] and its application to quantum optical scattering discussed in Ref. [75], suitably specialized here to a nonmagnetic object in vacuum, occupying a finite volume V and whose optical response is characterized in the frequency domain ($e^{-i\omega t}$) by the dielectric permittivity $\epsilon_\omega^V(\mathbf{s})$, which is a holomorphic function of ω for $\text{Im}(\omega) > 0$. We denote with $\epsilon_\omega(\mathbf{s})$ the overall permittivity coinciding with $\epsilon_\omega^V(\mathbf{s})$ within the object ($\mathbf{s} \in V$) and being equal to 1 in vacuum ($\mathbf{s} \notin V$), as depicted in Fig. 1(a). According to the MLNF, the quantization of the electromagnetic field in the presence of the lossy object is governed by the Hamiltonian operator

$$\hat{H} = \int d\omega \hbar \omega \left[\int d\mathbf{o}_n \sum_\lambda \hat{a}_s^\dagger(\omega \mathbf{n} \lambda) \hat{a}_s(\omega \mathbf{n} \lambda) + \int d^3 \mathbf{r} \sum_v \hat{a}_e^\dagger(\omega \mathbf{r} v) \hat{a}_e(\omega \mathbf{r} v) \right], \quad (1)$$

where $\hat{a}_s(\omega \mathbf{n} \lambda)$ and $\hat{a}_e(\omega \mathbf{r} v)$ are the annihilation operators for the s and e polaritons, respectively. These operators satisfy the standard bosonic commutation relations

$$[\hat{a}_s(\omega \mathbf{n} \lambda), \hat{a}_s^\dagger(\omega' \mathbf{n}' \lambda')] = \delta(\omega - \omega') \delta(\mathbf{o}_n - \mathbf{o}_{n'}) \delta_{\lambda \lambda'},$$

$$[\hat{a}_e(\omega \mathbf{r} v), \hat{a}_e^\dagger(\omega' \mathbf{r}' v')] = \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}') \delta_{vv'}, \quad (2)$$

with all other commutators vanishing. The s polaritons represent the field excitations (counterparts of photons in the lossy regime), while the e polaritons represent the quanta of the medium's dipolar sources. To simplify the readability of the paper and focus on the physical results, the detailed mathematical derivation of the scattering equations—which involves the classical modal dyadics, the dyadic Green's function, and the integration over the medium's degrees of freedom—is relegated to Appendix A. By applying this first-principle procedure, the quantum optical scattering process can be fully mapped onto a unitary input-output transformation connecting the ingoing polariton operators $\hat{a}_s(\omega \mathbf{n} \lambda)$ and $\hat{a}_e(\omega \mathbf{r} v)$ in the far past ($t \rightarrow -\infty$) to the outgoing polariton operators $\hat{A}_s(\omega \mathbf{N} \Lambda)$ and $\hat{A}_e(\omega \mathbf{R} \Upsilon)$ in the far future ($t \rightarrow +\infty$):

$$\begin{aligned} \hat{A}_s(\omega \mathbf{N} \Lambda) &= \int d\mathbf{o}_n \sum_\lambda [\mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathcal{T}_{oss}(\mathbf{N}|\mathbf{n}) \cdot \mathbf{e}_{\mathbf{n}\lambda}] \hat{a}_s(\omega \mathbf{n} \lambda) \\ &\quad + \int d^3 \mathbf{r} \sum_v [\mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathcal{E}_{ose}(\mathbf{N}|\mathbf{r}) \cdot \mathbf{u}_v] \hat{a}_e(\omega \mathbf{r} v), \\ \hat{A}_e(\omega \mathbf{R} \Upsilon) &= \int d\mathbf{o}_n \sum_\lambda [\mathbf{u}_\Upsilon \cdot \mathcal{A}_{oes}(\mathbf{R}|\mathbf{n}) \cdot \mathbf{e}_{\mathbf{n}\lambda}] \hat{a}_s(\omega \mathbf{n} \lambda) \\ &\quad + \int d^3 \mathbf{r} \sum_v [\mathbf{u}_\Upsilon \cdot \mathcal{Q}_{oee}(\mathbf{R}|\mathbf{r}) \cdot \mathbf{u}_v] \hat{a}_e(\omega \mathbf{r} v). \end{aligned} \quad (3)$$

Here, the linear mixture of the operators is governed by the macroscopic transmission ($\mathcal{T}_{oss}$), emission ($\mathcal{E}_{ose}$), absorption ($\mathcal{A}_{oes}$), and energy redistribution ($\mathcal{Q}_{oee}$) classical dyadics, which are explicitly defined in Appendix A. These outgoing operators play the exact same physical role in the far future as the ingoing ones do in the far past, since they represent the asymptotic output states of the scattering process. This is conceptually equivalent to the standard input-output relations describing lossy four-port devices [38–40], here generalized to a continuous macroscopic scatterer. This input-output transformation ensures that the bosonic commutation relations are conserved for the outgoing operators ($\hat{A}_s, \hat{A}_e$).

III. SCATTERING OF TWO DIRECTIONAL s POLARITONS

We here exploit the framework reviewed in the last section to analyze the quantum scattering of two noncollinear ingoing s polaritons by an electromagnetically inert lossy object (i.e., not hosting ingoing e polaritons).

A. Input and output radiation-matter descriptions

In the far past $t = -\infty$, the radiation-matter description is provided by the operators $\hat{a}_s$ and $\hat{a}_e$ so that, in order to put no restrictions on the frequency-polarization correlations between the two s polaritons, we assume that the Heisenberg state of the ingoing wave packet is

$$|\Psi\rangle = \int d\omega^1 d\omega^2 \int d\mathbf{o}_n^1 d\mathbf{o}_n^2 \sum_{\lambda^1 \lambda^2} \phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2) \Theta_{\mathbf{m}^1}(\mathbf{n}^1) \times \Theta_{\mathbf{m}^2}(\mathbf{n}^2) \hat{a}_s^\dagger(\omega^1 \mathbf{n}^1 \lambda^1) \hat{a}_s^\dagger(\omega^2 \mathbf{n}^2 \lambda^2) |0\rangle, \quad (4)$$

where $\mathbf{m}^1$ and $\mathbf{m}^2$ are unit vectors with $\mathbf{m}^1 \neq \mathbf{m}^2$, $\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)$ is a frequency-polarization spectrum and the function $\Theta_{\mathbf{m}}(\mathbf{n}) = \frac{1}{\sqrt{\Delta o}} H(\frac{\Delta \theta_n}{2} - |\theta_n - \theta_m|) H(\frac{\Delta \varphi_n}{2} - |\varphi_n - \varphi_m|)$, where H is the Heaviside step function, selects the unit vectors $\mathbf{n}$ within a fixed and very small solid angle $\Delta o = \Delta \theta_n \Delta \varphi_n \sin \theta_n \ll 1$ around the unit vector $\mathbf{m}$, and it is such that $\int d\mathbf{o}_n [\Theta_{\mathbf{m}}(\mathbf{n})]^2 = 1$. Accordingly, with reference to Fig. 1(a), the state in Eq. (4) is the superposition of all the basis states with two s polaritons whose directions $\mathbf{n}^1$ and $\mathbf{n}^2$ lie within the two thin cones (gray) around the directions $\mathbf{m}^1$ and $\mathbf{m}^2$ (red). We hereafter assume that such two cones do not overlap, i.e., $\Theta_{\mathbf{m}^1}(\mathbf{n}) \Theta_{\mathbf{m}^2}(\mathbf{n}) = 0$, so that the state normalization $\langle \Psi | \Psi \rangle = 1$ is easily seen to be assured by the spectral normalization

$$\int d\omega^1 d\omega^2 \sum_{\lambda^1 \lambda^2} |\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)|^2 = 1. \quad (5)$$

The ingoing polariton wave function $\Psi_{ss}^{(in)}(\omega^1 \mathbf{n}^1 \lambda^1, \omega^2 \mathbf{n}^2 \lambda^2) = \frac{1}{\sqrt{2}} \langle 0 | \hat{a}_s(\omega^1 \mathbf{n}^1 \lambda^1) \hat{a}_s(\omega^2 \mathbf{n}^2 \lambda^2) | \Psi \rangle$ is given by

$$\begin{aligned} \Psi_{ss}^{(in)}(\omega^1 \mathbf{n}^1 \lambda^1, \omega^2 \mathbf{n}^2 \lambda^2) &= \frac{1}{\sqrt{2}} [\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2) \Theta_{\mathbf{m}^1}(\mathbf{n}^1) \Theta_{\mathbf{m}^2}(\mathbf{n}^2) \\ &\quad + \phi_{\lambda^2 \lambda^1}(\omega^2, \omega^1) \Theta_{\mathbf{m}^1}(\mathbf{n}^2) \Theta_{\mathbf{m}^2}(\mathbf{n}^1)] \end{aligned} \quad (6)$$

and it is evidently symmetric under the exchange $\omega^1 \mathbf{n}^1 \lambda^1 \leftrightarrow \omega^2 \mathbf{n}^2 \lambda^2$ of the two s polaritons. It is worth pointing out that the noncollinearity of two thin cones of Fig. 1(a) $\mathbf{m}^1 \neq \mathbf{m}^2$ causes the wave function $\Psi_{ss}^{(in)}$ to be the sum of a direct and an exchange term whose respective amplitudes $\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)$ and $\phi_{\lambda^2 \lambda^1}(\omega^2, \omega^1)$ are generally different since the exchange $\omega^1 \lambda^1 \leftrightarrow \omega^2 \lambda^2$ does not amount to the physical exchange of the two s polaritons. In other words, the spectrum $\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)$ is not required to be exchange-symmetric since the s polaritons whose directions lie within the cones of Fig. 1(a) are distinguishable.

In the far future $t = +\infty$, the radiation-matter description is provided by the operators $\hat{A}_s$ and $\hat{A}_e$ and it can be obtained by eliminating the two creation operators $\hat{a}_s^\dagger$ in the state $|\Psi\rangle$ of Eq. (4) by means of the Hermitian conjugate of the first of Eq. (A11), which is

$$\begin{aligned} \hat{a}_s^\dagger(\omega \mathbf{n} \lambda) &= \int d\mathbf{o}_N \sum_{\Lambda} [\mathbf{e}_{N\Lambda} \cdot \mathcal{T}_{oss}(\mathbf{N}|\mathbf{n}) \cdot \mathbf{e}_{n\lambda}] \hat{A}_s^\dagger(\omega \mathbf{N} \Lambda) \\ &\quad + \int d^3 \mathbf{R} \sum_{\Upsilon} [\mathbf{u}_\Upsilon \cdot \mathcal{A}_{oes}(\mathbf{R}|\mathbf{n}) \cdot \mathbf{e}_{n\lambda}] \hat{A}_e^\dagger(\omega \mathbf{R} \Upsilon). \end{aligned} \quad (7)$$

The ensuing expression of the state displays three contributions containing $\hat{A}_s^\dagger \hat{A}_s^\dagger$, $\hat{A}_s^\dagger \hat{A}_e^\dagger$, and $\hat{A}_e^\dagger \hat{A}_e^\dagger$, respectively corresponding to the three physical processes where both s polaritons are scattered ($ss \rightarrow ss$), one is scattered and the other one decays into an e polariton ($ss \rightarrow se$), and both decay into two e polaritons ($ss \rightarrow ee$), as diagrammatically represented in Fig. 1(b). The amplitudes of the three processes are the two-photon survival wave function $\Psi_{ss}^{(out)}(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{N}^2 \Lambda^2) = \frac{1}{\sqrt{2}} \langle 0 | \hat{A}_s(\omega^1 \mathbf{N}^1 \Lambda^1) \hat{A}_s(\omega^2 \mathbf{N}^2 \Lambda^2) | \Psi \rangle$, the one-photon survival wave function $\Psi_{se}^{(out)}(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{R}^2 \Upsilon^2) = \langle 0 | \hat{A}_s(\omega^1 \mathbf{N}^1 \Lambda^1) \hat{A}_e(\omega^2 \mathbf{R}^2 \Upsilon^2) | \Psi \rangle$, and the zero-photon survival wave function $\Psi_{ee}^{(out)}(\omega^1 \mathbf{R}^1 \Upsilon^1, \omega^2 \mathbf{R}^2 \Upsilon^2) = \frac{1}{\sqrt{2}} \langle 0 | \hat{A}_e(\omega^1 \mathbf{R}^1 \Upsilon^1) \hat{A}_e(\omega^2 \mathbf{R}^2 \Upsilon^2) | \Psi \rangle$. In order not to overburden the mathematical treatment, we resort to the following three simplifying, but not very restrictive, assumptions.

(1) We omit the delta function contribution in the transmission dyadic $\mathcal{T}_{oss}(\mathbf{N}|\mathbf{n})$ of the first of Eq. (A7); this corresponds to analyzing the only scattered field (i.e., without the incident plane wave field contribution). This assumption is strictly valid for outgoing s polaritons whose directions $\mathbf{N}^1$ and $\mathbf{N}^2$ do not lie within the two thin cones of Fig. 1(a), i.e., $\Theta_{\mathbf{m}^1}(\mathbf{N}^1) = \Theta_{\mathbf{m}^2}(\mathbf{N}^1) = 0$ and $\Theta_{\mathbf{m}^1}(\mathbf{N}^2) = \Theta_{\mathbf{m}^2}(\mathbf{N}^2) = 0$.

(2) We assume that the scattering and absorption dyadics $\mathcal{S}_\omega(\mathbf{N}|\mathbf{n})$ and $\mathcal{A}_{oes}(\mathbf{R}|\mathbf{n})$ do not appreciably vary when $\mathbf{n}$ spans the small solid angle Δo in such a way that the approximation $\int d\mathbf{o}_n \Theta_{\mathbf{m}}(\mathbf{n}) f(\mathbf{n}) \simeq \sqrt{\Delta o} f(\mathbf{m})$ can be safely used.

(3) We assume that the frequency bandwidth of the spectrum $\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)$ is narrow enough that the frequency dependence of $\mathcal{S}_\omega(\mathbf{N}|\mathbf{n})$ and $\mathcal{A}_{oes}(\mathbf{R}|\mathbf{n})$ can be neglected over the bandwidth, so that they can be evaluated at a central bandwidth frequency ω .

Under the above assumptions, after tedious but straightforward algebra, the outgoing wave functions are seen to be

given by

$$\begin{aligned}
& \Psi_{ss}^{(\text{out})}(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{N}^2 \Lambda^2) \\
&= \frac{\Delta o}{\sqrt{2}} \sum_{\lambda^1 \lambda^2} [\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2) s_{\mathbf{m}^1 \lambda^1}^{\mathbf{N}^1 \Lambda^1} s_{\mathbf{m}^2 \lambda^2}^{\mathbf{N}^2 \Lambda^2} \\
&\quad + \phi_{\lambda^1 \lambda^2}(\omega^2, \omega^1) s_{\mathbf{m}^1 \lambda^1}^{\mathbf{N}^2 \Lambda^2} s_{\mathbf{m}^2 \lambda^2}^{\mathbf{N}^1 \Lambda^1}], \\
& \Psi_{se}^{(\text{out})}(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{R}^2 \Upsilon^2) \\
&= \Delta o \sum_{\lambda^1 \lambda^2} [\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2) s_{\mathbf{m}^1 \lambda^1}^{\mathbf{N}^1 \Lambda^1} a_{\mathbf{m}^2 \lambda^2}^{\mathbf{R}^2 \Upsilon^2} \\
&\quad + \phi_{\lambda^1 \lambda^2}(\omega^2, \omega^1) a_{\mathbf{m}^1 \lambda^1}^{\mathbf{R}^2 \Upsilon^2} s_{\mathbf{m}^2 \lambda^2}^{\mathbf{N}^1 \Lambda^1}], \\
& \Psi_{ee}^{(\text{out})}(\omega^1 \mathbf{R}^1 \Upsilon^1, \omega^2 \mathbf{R}^2 \Upsilon^2) \\
&= \frac{\Delta o}{\sqrt{2}} \sum_{\lambda^1 \lambda^2} [\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2) a_{\mathbf{m}^1 \lambda^1}^{\mathbf{R}^1 \Upsilon^1} a_{\mathbf{m}^2 \lambda^2}^{\mathbf{R}^2 \Upsilon^2} \\
&\quad + \phi_{\lambda^1 \lambda^2}(\omega^2, \omega^1) a_{\mathbf{m}^1 \lambda^1}^{\mathbf{R}^2 \Upsilon^2} a_{\mathbf{m}^2 \lambda^2}^{\mathbf{R}^1 \Upsilon^1}], \tag{8}
\end{aligned}$$

where

$$\begin{aligned}
s_{\mathbf{n}\lambda}^{\mathbf{N}\Lambda}(\omega) &= \mathbf{e}_{\mathbf{n}\Lambda} \cdot \frac{ik_\omega}{2\pi} \mathcal{S}_\omega(\mathbf{N}|\mathbf{n}) \cdot \mathbf{e}_{\mathbf{n}\lambda}, \\
a_{\mathbf{n}\lambda}^{\mathbf{R}\Upsilon}(\omega) &= \mathbf{u}_\Upsilon \cdot \mathcal{A}_{\omega\epsilon\mathbf{s}}(\mathbf{R}|\mathbf{n}) \cdot \mathbf{e}_{\mathbf{n}\lambda} \tag{9}
\end{aligned}$$

are the scattering and absorption coefficients of the scatterer object. The wave functions $\Psi_{ss}^{(\text{out})}$ and $\Psi_{ee}^{(\text{out})}$ evidently display the exchange symmetries $\omega^1 \mathbf{N}^1 \Lambda^1 \leftrightarrow \omega^2 \mathbf{N}^2 \Lambda^2$ and $\omega^1 \mathbf{R}^1 \Upsilon^1 \leftrightarrow \omega^2 \mathbf{R}^2 \Upsilon^2$, respectively. The crucial point here is that each of the three wave functions in Eq. (8) is the sum of a direct and an exchange contribution, respectively proportional to the amplitudes $\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)$ and $\phi_{\lambda^2 \lambda^1}(\omega^2, \omega^1)$ (after a $\lambda^1 \lambda^2$ relabelling) appearing in the ingoing wave function of Eq. (6). Moreover, the subscripts and superscripts of the coefficients $s_{\mathbf{n}\lambda}^{\mathbf{N}\Lambda}$ and $a_{\mathbf{n}\lambda}^{\mathbf{R}\Upsilon}$ in Eq. (8) reveal that, in each process, the direct and exchange contributions correspond to the two possible quantum paths connecting the ingoing s polaritons $\omega^1 \mathbf{m}^1 \lambda^1$ and $\omega^2 \mathbf{m}^2 \lambda^2$ to the two outgoing ones involved in the process, as graphically sketched in Fig. 1(c). Therefore, we conclude that the detection probability of each of the three processes can display quantum interference effects arising from the interplay between the direct and exchange paths, in turn resulting from the ingoing s polaritons' noncollinearity [assigning distinct physical meanings to $\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)$ and $\phi_{\lambda^2 \lambda^1}(\omega^2, \omega^1)$ in Eq. (6)] and from electromagnetic propagation [provided by the classical scattering and absorption dyadics $\mathcal{S}_\omega(\mathbf{N}|\mathbf{n})$ and $\mathcal{A}_{\omega\epsilon\mathbf{s}}(\mathbf{R}|\mathbf{n})$].

B. Wave packets with definite spectral symmetry

From Eq. (8), it is manifest that, for fixed directions $\mathbf{m}^1$ and $\mathbf{m}^2$, the ingoing wave packets producing the largest interference effects are those whose spectrum is such that $|\phi_{\lambda^2 \lambda^1}(\omega^2, \omega^1)| = |\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2)|$, so that we hereafter focus on ingoing wave packets whose spectrum $\phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2)$ is either symmetric ($\sigma = 1$) or antisymmetric ($\sigma = -1$), i.e.,

$$\phi_{\lambda^2 \lambda^1}^\sigma(\omega^2, \omega^1) = \sigma \phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2), \tag{10}$$

a condition which also considerably simplifies the mathematical treatment [79]. From Eqs. (10) and (6), the wave function corresponding to the spectrum $\phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2)$ is

$$\begin{aligned}
& \Psi_{ss}^{(\text{in})\sigma}(\omega^1 \mathbf{n}^1 \lambda^1, \omega^2 \mathbf{n}^2 \lambda^2) \\
&= \frac{1}{\sqrt{2}} \phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2) [\Theta_{\mathbf{m}^1}(\mathbf{n}^1) \Theta_{\mathbf{m}^2}(\mathbf{n}^2) \\
&\quad + \sigma \Theta_{\mathbf{m}^1}(\mathbf{n}^2) \Theta_{\mathbf{m}^2}(\mathbf{n}^1)], \tag{11}
\end{aligned}$$

so that the exchange $\mathbf{m}^1 \leftrightarrow \mathbf{m}^2$ yields the wave-function change $\Psi_{ss}^{(\text{in})\sigma} \rightarrow \sigma \Psi_{ss}^{(\text{in})\sigma}$, i.e., it leaves invariant the probability density. Physically, this means that an ingoing wave packet with symmetric or antisymmetric spectrum is such that the flows of its s polaritons through the two cones around the fixed directions $\mathbf{m}^1$ and $\mathbf{m}^2$ shown in Fig. 1(a) are identical. It is worth noting that the ingoing s polaritons with antisymmetric spectrum are necessarily entangled since a separable spectrum $\phi_{\lambda^1 \lambda^2}(\omega^1, \omega^2) = \xi_{\lambda^1}(\omega^1) \zeta_{\lambda^2}(\omega^2)$ pertaining to not-entangled s polaritons cannot be antisymmetric; accordingly, the possible detection of interference effects pertinent to the spectral antisymmetry is a distinct signature of the ingoing s polariton entanglement.

Substituting Eq. (10) into Eq. (8) and taking the square moduli, we get the outgoing polariton joint detection probability densities:

$$\begin{aligned}
& \frac{dP_{ss}^\sigma(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{N}^2 \Lambda^2)}{d\omega^1 d\mathbf{o}_{\mathbf{N}^1} d\omega^2 d\mathbf{o}_{\mathbf{N}^2}} \\
&= \frac{\Delta o^2}{2} \left| \sum_{\lambda^1 \lambda^2} \phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2) (s_{\mathbf{m}^1 \lambda^1}^{\mathbf{N}^1 \Lambda^1} s_{\mathbf{m}^2 \lambda^2}^{\mathbf{N}^2 \Lambda^2} + \sigma s_{\mathbf{m}^1 \lambda^2}^{\mathbf{N}^2 \Lambda^2} s_{\mathbf{m}^2 \lambda^1}^{\mathbf{N}^1 \Lambda^1}) \right|^2, \\
& \frac{dP_{se}^\sigma(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{R}^2 \Upsilon^2)}{d\omega^1 d\mathbf{o}_{\mathbf{N}^1} d\omega^2 d^3 \mathbf{R}^2} \\
&= \Delta o^2 \left| \sum_{\lambda^1 \lambda^2} \phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2) (s_{\mathbf{m}^1 \lambda^1}^{\mathbf{N}^1 \Lambda^1} a_{\mathbf{m}^2 \lambda^2}^{\mathbf{R}^2 \Upsilon^2} + \sigma a_{\mathbf{m}^1 \lambda^2}^{\mathbf{R}^2 \Upsilon^2} s_{\mathbf{m}^2 \lambda^1}^{\mathbf{N}^1 \Lambda^1}) \right|^2, \\
& \frac{dP_{ee}^\sigma(\omega^1 \mathbf{R}^1 \Upsilon^1, \omega^2 \mathbf{R}^2 \Upsilon^2)}{d\omega^1 d^3 \mathbf{R}^1 d\omega^2 d^3 \mathbf{R}^2} \\
&= \frac{\Delta o}{2} \left| \sum_{\lambda^1 \lambda^2} \phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2) (a_{\mathbf{m}^1 \lambda^1}^{\mathbf{R}^1 \Upsilon^1} a_{\mathbf{m}^2 \lambda^2}^{\mathbf{R}^2 \Upsilon^2} + \sigma a_{\mathbf{m}^1 \lambda^2}^{\mathbf{R}^2 \Upsilon^2} a_{\mathbf{m}^2 \lambda^1}^{\mathbf{R}^1 \Upsilon^1}) \right|^2, \tag{12}
\end{aligned}$$

whose associated complete scattering description is rather involved due to the number of polariton variables. To simplify the analysis, bearing in mind that outgoing e polaritons are usually left unmeasured in standard setup (i.e., no measurement is performed on the object), we focus on the processes where two, one, and zero outgoing s polaritons are globally scattered whose probabilities, namely, the two-polariton survival probability P_{2s}^σ , the single-polariton survival probability P_{1s}^σ , and the complete absorption probability P_{0s}^σ , in view of

the first assumption of Sec. III A, are given by

$$\begin{aligned}
 P_{2s}^\sigma &= \int d\omega^1 \int d\omega_{N^1} \sum_{\Lambda^1} \int d\omega^2 \int d\omega_{N^2} \sum_{\Lambda^2} \\
 &\quad \times \frac{dP_{ss}^\sigma(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{N}^2 \Lambda^2)}{d\omega^1 d\omega_{N^1} d\omega^2 d\omega_{N^2}}, \\
 P_{1s}^\sigma &= \int d\omega^1 \int d\omega_{N^1} \sum_{\Lambda^1} \int d\omega^2 \int d^3 \mathbf{R}^2 \sum_{\Upsilon^2} \\
 &\quad \times \frac{dP_{se}^\sigma(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{R}^2 \Upsilon^2)}{d\omega^1 d\omega_{N^1} d\omega^2 d^3 \mathbf{R}^2}, \\
 P_{0s}^\sigma &= \int d\omega^1 \int d^3 \mathbf{R}^1 \sum_{\Upsilon^1} \int d\omega^2 \int d^3 \mathbf{R}^2 \sum_{\Upsilon^2} \\
 &\quad \times \frac{dP_{ee}^\sigma(\omega^1 \mathbf{R}^1 \Upsilon^1, \omega^2 \mathbf{R}^2 \Upsilon^2)}{d\omega^1 d^3 \mathbf{R}^1 d\omega^2 d^3 \mathbf{R}^2}. \quad (13)
 \end{aligned}$$

Note that, in the angular integrations of Eq. (13), we have included the outgoing s polariton directions $\mathbf{N}^1$ and $\mathbf{N}^2$ belonging to the incident cones of Fig. 1(a) (which are excluded by the above first assumption of Sec. III A) by committing a negligible error since $\Delta o \ll 1$. After some algebra, the scattering probabilities of Eq. (13) turn out to be

$$\begin{aligned}
 P_{2s}^\sigma &= \Delta o^2 [\langle S_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} \rangle^\sigma + \sigma \langle S_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} \rangle^\sigma], \\
 P_{1s}^\sigma &= \Delta o^2 [\langle S_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} A_{\mathbf{m}^2 \lambda^2}^{\mathbf{m}^1 \lambda^1} + A_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} \rangle^\sigma \\
 &\quad + \sigma \langle S_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} A_{\mathbf{m}^2 \lambda^2}^{\mathbf{m}^1 \lambda^1} + A_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} \rangle^\sigma], \\
 P_{0s}^\sigma &= \Delta o^2 [\langle A_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} A_{\mathbf{m}^2 \lambda^2}^{\mathbf{m}^1 \lambda^1} \rangle^\sigma + \sigma \langle A_{\mathbf{m}^1 \lambda^1}^{\mathbf{m}^2 \lambda^2} A_{\mathbf{m}^2 \lambda^2}^{\mathbf{m}^1 \lambda^1} \rangle^\sigma], \quad (14)
 \end{aligned}$$

where

$$\begin{aligned}
 S_{\mathbf{n} \lambda'}^{\mathbf{n} \lambda}(\omega) &= \mathbf{e}_{\mathbf{n} \lambda} \cdot \left[\left(\frac{k_\omega}{2\pi} \right)^2 \int d\omega_{\mathbf{N}} S_\omega^{T*}(\mathbf{N}|\mathbf{n}) \cdot S_\omega(\mathbf{N}|\mathbf{n}') \right] \cdot \mathbf{e}_{\mathbf{n} \lambda'}, \\
 A_{\mathbf{n} \lambda'}^{\mathbf{n} \lambda}(\omega) &= \mathbf{e}_{\mathbf{n} \lambda} \cdot \left[\int d^3 \mathbf{R} \mathcal{A}_{\omega es}^{T*}(\mathbf{R}|\mathbf{n}) \cdot \mathcal{A}_{\omega es}(\mathbf{R}|\mathbf{n}') \right] \cdot \mathbf{e}_{\mathbf{n} \lambda'} \quad (15)
 \end{aligned}$$

are the dimensionless power scattering and absorption coefficients of the scatterer object satisfying the Hermiticity relations $(S_{\mathbf{n} \lambda'}^{\mathbf{n} \lambda})^* = S_{\mathbf{n} \lambda}^{\mathbf{n} \lambda'}$ and $(A_{\mathbf{n} \lambda'}^{\mathbf{n} \lambda})^* = A_{\mathbf{n} \lambda}^{\mathbf{n} \lambda'}$, whereas

$$\begin{aligned}
 \langle Q_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2} \rangle^\sigma &= \sum_{\lambda^1 \lambda^2 \lambda^1 \lambda^2} (I^\sigma)_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2} Q_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2}, \\
 (I^\sigma)_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2} &= \int d\omega^1 d\omega^2 \phi_{\lambda^1 \lambda^2}^{\sigma*}(\omega^1, \omega^2) \phi_{\lambda^1 \lambda^2}^\sigma(\omega^1, \omega^2) \quad (16)
 \end{aligned}$$

is the average of the classical quantity Q performed by means of the spectral overlap integrals I^σ of the ingoing wave packet spectrum, in turn satisfying the relations [see Eqs. (5) and (10)]

$$\begin{aligned}
 \sum_{\lambda^1 \lambda^2} (I^\sigma)_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2} &= 1, \quad [(I^\sigma)_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2}]^* = (I^\sigma)_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2}, \\
 (I^\sigma)_{\lambda^2 \lambda^1}^{\lambda^2 \lambda^1} &= (I^\sigma)_{\lambda^1 \lambda^2}^{\lambda^1 \lambda^2}. \quad (17)
 \end{aligned}$$

Note that three probabilities in Eq. (14) have two contributions, respectively characterized by their $\mathbf{m}^1 \mathbf{m}^2$ label placements $(\mathbf{m}^1 \mathbf{m}^2)$ and $(\mathbf{m}^2 \mathbf{m}^1)$, the second of which has the

factor σ . From all the above discussion, it follows that the $(\mathbf{m}^2 \mathbf{m}^1)$ terms are responsible for the quantum interference effects showing up in the s polariton scattering probabilities and that such interference is affected by the symmetry σ of the ingoing wave-packet spectrum.

C. Physical meaning of power scattering and absorption coefficients

Before undertaking the analysis of the wave packet quantum scattering by a sphere, we here discuss the physical meaning of power scattering and absorption coefficients in Eq. (15). To this end, we consider the classical scattering by an arbitrary lossy object of the incident field $\mathbf{E}_\omega^{(\text{in})}(\mathbf{s}) = E_0 \sum_{i=1}^N e^{i\mathbf{k}_\omega \mathbf{m}^i \cdot \mathbf{s}} \mathbf{e}^i$, which is the superposition of N plane waves with equal amplitude E_0 , directions $\mathbf{m}^i$ and polarizations $\mathbf{e}^i$ (with $\mathbf{m}^i \cdot \mathbf{m}^i = 1$, $\mathbf{e}^i \cdot \mathbf{e}^{i*} = 1$, and $\mathbf{e}^i \cdot \mathbf{m}^i = 0$). A simple extension of the single plane wave scattering analysis (i.e., see Ref. [80]) enables us to prove that the N -plane scattering is characterized by the scattering, extinction, and absorption cross sections given by

$$\begin{aligned}
 C_{\text{sca}} &= \sum_{ij} \mathbf{e}^{i*} \cdot \left[\int d\omega_{\mathbf{N}} S_\omega^{T*}(\mathbf{N}|\mathbf{m}^i) \cdot S_\omega(\mathbf{N}|\mathbf{m}^j) \right] \cdot \mathbf{e}^j, \\
 C_{\text{ext}} &= \sum_{ij} \mathbf{e}^{i*} \cdot \frac{2\pi}{ik_\omega} [S_\omega(\mathbf{m}^i|\mathbf{m}^j) - S_\omega^{T*}(\mathbf{m}^j|\mathbf{m}^i)] \cdot \mathbf{e}^j, \\
 C_{\text{abs}} &= C_{\text{ext}} - C_{\text{sca}}, \quad (18)
 \end{aligned}$$

where each cross section is the ratio between the corresponding power and the intensity $|E_0|^2/(2\mu_0 c)$ of each plane wave. It is essential now to observe that, using the transmission dyadic $\mathcal{T}_{\omega ss}(\mathbf{N}|\mathbf{n})$ in the first of Eq. (A7), the first of Eq. (A8) turns into

$$\begin{aligned}
 &\int d^3 \mathbf{R} \mathcal{A}_{\omega es}^{T*}(\mathbf{R}|\mathbf{n}) \cdot \mathcal{A}_{\omega es}(\mathbf{R}|\mathbf{n}') \\
 &= \left(\frac{k_\omega}{2\pi} \right)^2 \left\{ \frac{2\pi}{ik_\omega} [S_\omega(\mathbf{n}|\mathbf{n}') - S_\omega^{T*}(\mathbf{n}'|\mathbf{n})] \right. \\
 &\quad \left. - \int d\omega_{\mathbf{N}} S_\omega^{T*}(\mathbf{N}|\mathbf{n}) \cdot S_\omega(\mathbf{N}|\mathbf{n}') \right\}, \quad (19)
 \end{aligned}$$

whose straightforward consequence is that the absorption cross section can be rewritten as

$$\begin{aligned}
 C_{\text{abs}} &= \left(\frac{2\pi}{k_\omega} \right)^2 \sum_{ij} \mathbf{e}^{i*} \cdot \left[\int d^3 \mathbf{R} \mathcal{A}_{\omega es}^{T*}(\mathbf{R}|\mathbf{m}^i) \cdot \mathcal{A}_{\omega es}(\mathbf{R}|\mathbf{m}^j) \right] \\
 &\quad \cdot \mathbf{e}^j. \quad (20)
 \end{aligned}$$

Eventually, by expanding the polarizations $\mathbf{e}^i$ of the plane waves on the hereafter considered basis $\{\mathbf{e}_{\mathbf{m}^i 1}, \mathbf{e}_{\mathbf{m}^i 2}\}$, i.e., setting $\mathbf{e}^i = \sum_\lambda c_\lambda^i \mathbf{e}_{\mathbf{m}^i \lambda}$, and using Eq. (15), the scattering and absorption cross section can be cast as

$$\begin{aligned}
 C_{\text{sca}} &= \left(\frac{2\pi}{k_\omega} \right)^2 \sum_{ij} \sum_{\lambda \lambda'} c_\lambda^{i*} c_{\lambda'}^j S_{\mathbf{m}^i \lambda'}^{\mathbf{m}^j \lambda}, \\
 C_{\text{abs}} &= \left(\frac{2\pi}{k_\omega} \right)^2 \sum_{ij} \sum_{\lambda \lambda'} c_\lambda^{i*} c_{\lambda'}^j A_{\mathbf{m}^i \lambda'}^{\mathbf{m}^j \lambda}, \quad (21)
 \end{aligned}$$

expressions elucidating the physical meaning of the power scattering and absorption coefficients. Specifically, apart from the numerical factor $(2\pi/k_\omega)^2$, the diagonal terms $S_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}$ and $A_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}$ represent the usual scattering and absorption cross sections of the plane wave $e^{ik_\omega \mathbf{n} \cdot \mathbf{s}} \mathbf{e}_{\mathbf{n}\lambda}$, whereas the off-diagonal terms $S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ and $A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ represent the contributions of the interference between the plane waves $e^{ik_\omega \mathbf{n} \cdot \mathbf{s}} \mathbf{e}_{\mathbf{n}\lambda}$ and $e^{ik_\omega \mathbf{n}' \cdot \mathbf{s}} \mathbf{e}_{\mathbf{n}'\lambda'}$ to the overall scattering and absorption cross sections.

IV. HOMOGENEOUS LOSSY SPHERE

We now specialize the above general description of quantum scattering to the relevant and analytically treatable situation where the scatterer object is a homogeneous lossy sphere of radius a and uniform dielectric permittivity ε_ω^V . Since we will be mainly interested in analyzing the coincidence detection and survival probabilities of the outgoing s polaritons [see the First of Eq. (8) and Eq. (14)], we here discuss the scattering coefficient $s_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}$ in Eq. (9) and the power scattering and absorption coefficients $S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ and $A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ in Eq. (15) of a homogeneous sphere, which (as detailed in Appendix B) are given by

$$\begin{aligned} s_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}(\omega) &= \mathbf{J}_{\mathbf{N}, \mathbf{n}} \left\{ \begin{pmatrix} -\frac{1}{2}a_{n\omega} & -\frac{1}{2}b_{n\omega} \end{pmatrix} \right\} \begin{pmatrix} \mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathbf{e}_{\mathbf{n}\lambda} \\ \mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathbf{n} \cdot \mathbf{e}_{\mathbf{n}\lambda} \end{pmatrix}, \\ S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega) &= \mathbf{J}_{\mathbf{n}, \mathbf{n}'} \{ (|a_{n\omega}|^2 \quad |b_{n\omega}|^2) \} \begin{pmatrix} \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \\ \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{n}' \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \end{pmatrix}, \\ A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega) &= \mathbf{J}_{\mathbf{n}, \mathbf{n}'} \{ (\text{Re}(a_{n\omega}) - |a_{n\omega}|^2 \quad \text{Re}(b_{n\omega}) - |b_{n\omega}|^2) \} \\ &\quad \times \begin{pmatrix} \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \\ \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{n}' \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \end{pmatrix}, \end{aligned} \quad (22)$$

where, for any pair of complex sequences p_n and q_m , we have introduced the row vector

$$\begin{aligned} \mathbf{J}_\xi \{ (p_n \quad q_m) \} &= \frac{1}{\pi} \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (p_n \quad q_m) \\ &\quad \times \begin{pmatrix} P'_n(\xi) & P''_n(\xi) \\ -(1-\xi^2)P''_n(\xi) + \xi P'_n(\xi) & -\xi P''_n(\xi) - P'_n(\xi) \end{pmatrix}, \end{aligned} \quad (23)$$

where $P_n(\xi)$ is the Legendre polynomial and $a_{n\omega}, b_{n\omega}$ are the standard Mie coefficients pertaining to the scattering of a single plane wave by a homogeneous sphere [81], i.e.,

$$\begin{aligned} a_{n\omega} &= \frac{j_n(k_\omega^V a) \frac{\partial}{\partial a} [a j_n(k_\omega a)] - \frac{1}{\varepsilon_\omega^V} j_n(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)]}{j_n(k_\omega^V a) \frac{\partial}{\partial a} [a h_n^{(1)}(k_\omega a)] - \frac{1}{\varepsilon_\omega^V} h_n^{(1)}(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)]}, \\ b_{n\omega} &= \frac{j_n(k_\omega^V a) \frac{\partial}{\partial a} [a j_n(k_\omega a)] - j_n(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)]}{j_n(k_\omega^V a) \frac{\partial}{\partial a} [a h_n^{(1)}(k_\omega a)] - h_n^{(1)}(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)]}, \end{aligned} \quad (24)$$

where $k_\omega^V = k_\omega(\varepsilon_\omega^V)^{1/2}$ [with $\text{Im}(k_\omega^V) \geq 0$] is the wave number inside the sphere and $j_n(\rho)$ and $h_n^{(1)}(\rho)$ are the spherical Bessel and Hankel functions of the first kind.

Note that the direction $\mathbf{m} = \{\mathbf{N}, \mathbf{n}, \mathbf{n}'\}$ and polarization $\mathbf{e}_{\mathbf{m}\mu} = \{\mathbf{e}_{\mathbf{N}\Lambda}, \mathbf{e}_{\mathbf{n}\lambda}, \mathbf{e}_{\mathbf{n}'\lambda'}\}$ unit vectors appear in Eq. (22) only

inside scalar products, with the result that $s_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}$, $S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ and $A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ are left invariant by any proper or improper rotation of the whole scattering setup $\mathbf{m} \rightarrow \mathcal{R} \cdot \mathbf{m}$ and $\mathbf{e}_{\mathbf{m}\mu} \rightarrow \mathcal{R} \cdot \mathbf{e}_{\mathbf{m}\mu}$ (where $\mathcal{R}$ is an orthogonal dyadic, i.e., $\mathcal{R} \cdot \mathcal{R}^T = \mathcal{R}^T \cdot \mathcal{R} = \mathcal{I}$), an evident consequence of the sphere symmetry. From Eq. (22), the diagonal power scattering and absorption coefficients are

$$\begin{aligned} S_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}(\omega) &= \mathbf{J}_1 \{ (|a_{n\omega}|^2 \quad |b_{n\omega}|^2) \} \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \\ A_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}(\omega) &= \mathbf{J}_1 \{ (\text{Re}(a_{n\omega}) - |a_{n\omega}|^2 \quad \text{Re}(b_{n\omega}) - |b_{n\omega}|^2) \} \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \end{aligned} \quad (25)$$

so that, since $P'_n(1) = n(n+1)/2$, Eq. (23) directly yields

$$\begin{aligned} \left(\frac{2\pi}{k_\omega} \right)^2 S_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}(\omega) &= \frac{2\pi}{k_\omega^2} \sum_{n=1}^{\infty} (2n+1) (|a_{n\omega}|^2 + |b_{n\omega}|^2) \equiv C_{\text{sca}}^S, \\ \left(\frac{2\pi}{k_\omega} \right)^2 A_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}(\omega) &= \frac{2\pi}{k_\omega^2} \sum_{n=1}^{\infty} (2n+1) \{ [\text{Re}(a_{n\omega}) - |a_{n\omega}|^2] \\ &\quad + [\text{Re}(b_{n\omega}) - |b_{n\omega}|^2] \} \equiv C_{\text{abs}}^S, \end{aligned} \quad (26)$$

where C_{sca}^S and C_{abs}^S are the classical scattering and absorption cross sections pertaining to the scattering of any single plane wave $e^{ik_\omega \mathbf{n} \cdot \mathbf{s}} \mathbf{e}_{\mathbf{n}\lambda}$ by the sphere [81], in agreement with the discussion in Sec. III C. As a last remark on the coefficients in Eq. (22), it is worth discussing the two specific situations where there is no sphere $\varepsilon_\omega^V = 1$ or the sphere is ideally transparent $\text{Im}(\varepsilon_\omega^V) = 0$; they provide consistency checks of our treatment. In the first situation, the Mie coefficients $a_{n\omega}$ and $b_{n\omega}$ vanish [see Eq. (24)] and consequently, from Eq. (22), the coefficients $s_{\mathbf{n}\lambda}^{\mathbf{n}\lambda}$, $S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ and $A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ vanish as well, i.e., there is correctly no radiation scattering. In the second situation, as shown in Appendix B, the Mie coefficients are such that $\text{Re}(a_{n\omega}) = |a_{n\omega}|^2$ and $\text{Re}(b_{n\omega}) = |b_{n\omega}|^2$ so that, from the third of Eq. (22), the power absorption coefficient $A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}$ vanishes, i.e., the sphere correctly does not absorb energy from the radiation.

Having established the two s polariton scattering formalism for the realistic case of a homogeneous lossy sphere, the remainder of this work is dedicated to investigating two distinct features of this interference phenomenon. Specifically, Sec. V analyzes the spatial characteristics, identifying the precise scattering geometries required to achieve perfect interference in the polariton-coincidence detection. Subsequently, Sec. VI focuses on the spectral features of these quantum effects, exploring how they are governed by the frequency distribution of the impinging wave packets and the intrinsic material dissipation.

V. SCATTERING GEOMETRIES FOR PERFECT INTERFERENCE IN THE POLARITON-COINCIDENCE DETECTION

In addition to the overall rotational invariance mentioned in Sec. III D, we here show that the sphere symmetry entails the existence of specific scattering geometries such that, regardless of the wave-packet spectral profile, the probability

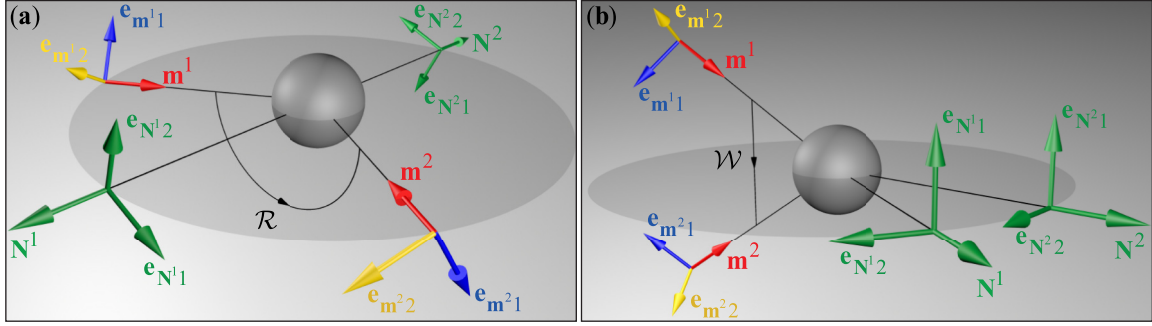


FIG. 2. Sample classes of scattering geometries leading to perfect interference in the polariton-coincidence detection. (a) Scattering geometry belonging to the first class, where the input ports are mutually related by a π rotation $\mathcal{R}$. This geometry yields perfect constructive or destructive interference for symmetric ($\sigma = 1$) or antisymmetric ($\sigma = -1$) input wave packets, respectively. (b) Scattering geometry belonging to the second class, where the input ports are mutually related by a reflection $\mathcal{W}$. Here, the resulting constructive or destructive interference depends on both the wave packet symmetry σ and the selected detection polarizations. In both panels, the outgoing polariton triads $\{\mathbf{N}^i, \mathbf{e}_{\mathbf{N}^i\lambda}, \mathbf{e}_{\mathbf{N}^i\lambda^2}\}$ ($i = 1, 2$), representing the detectors' spatial placements and polarization directions, are plotted in green. The ingoing polariton triads $\{\mathbf{m}^i, \mathbf{e}_{\mathbf{m}^i\lambda}, \mathbf{e}_{\mathbf{m}^i\lambda^2}\}$, representing the input ports through which light is launched, are plotted in distinct colors (red, blue, and yellow) to explicitly outline the mapping action of the geometric transformations $\mathcal{R}$ and $\mathcal{W}$.

density of polariton-coincidence detection displays perfect constructive or destructive interference.

With reference to the $ss \rightarrow ss$ process discussed in Sec. III A, a scattering geometry is assigned by the four orthonormal triads $\{\mathbf{m}^1, \mathbf{e}_{\mathbf{m}^1\lambda}, \mathbf{e}_{\mathbf{m}^1\lambda^2}\}$, $\{\mathbf{m}^2, \mathbf{e}_{\mathbf{m}^2\lambda}, \mathbf{e}_{\mathbf{m}^2\lambda^2}\}$, $\{\mathbf{N}^1, \mathbf{e}_{\mathbf{N}^1\lambda}, \mathbf{e}_{\mathbf{N}^1\lambda^2}\}$, $\{\mathbf{N}^2, \mathbf{e}_{\mathbf{N}^2\lambda}, \mathbf{e}_{\mathbf{N}^2\lambda^2}\}$, i.e., by the directions and polarizations of the ingoing and outgoing s polaritons. In other words, a scattering geometry represents the geometric arrangement in space of the “input ports” through which light is launched and of the “output ports” hosting the detectors, regardless of the spectrum ϕ^σ accounting for the quantum correlations between the ingoing s polaritons. Now the first of Eq. (12) shows that the probability density of polariton-coincidence detection depends on the scattering geometry only through the terms $s_{\mathbf{m}^1\lambda^1}^{\mathbf{N}^1\Lambda^1} s_{\mathbf{m}^2\lambda^2}^{\mathbf{N}^2\Lambda^2}$ and $s_{\mathbf{m}^1\lambda^2}^{\mathbf{N}^1\Lambda^2} s_{\mathbf{m}^2\lambda^1}^{\mathbf{N}^1\Lambda^1}$ representing the electromagnetic contributions to the two quantum paths involved in the $ss \rightarrow ss$ process, as discussed in Sec. III A. Therefore, a geometric strategy to achieve perfect interference involves exploiting the spherical symmetry to make the two quantum paths fully equivalent, or, in other words, to identify the scattering geometries such that the relation

$$\sum_{\lambda^1\lambda^2} \phi_{\lambda^1\lambda^2}^\sigma(\omega^1, \omega^2) s_{\mathbf{m}^1\lambda^2}^{\mathbf{N}^1\Lambda^2} s_{\mathbf{m}^2\lambda^1}^{\mathbf{N}^1\Lambda^1} = \pm \sum_{\lambda^1\lambda^2} \phi_{\lambda^1\lambda^2}^\sigma(\omega^1, \omega^2) s_{\mathbf{m}^1\lambda^1}^{\mathbf{N}^1\Lambda^1} s_{\mathbf{m}^2\lambda^2}^{\mathbf{N}^2\Lambda^2} \quad (27)$$

holds. Accordingly, Eq. (27) causes the probability density of polariton-coincidence detection in the first of Eq. (12) to have the overall factor $(1 \pm \sigma)^2$ (see below) so that perfect interference is achieved independently of the spectrum frequency profile. Due to the high symmetry of the sphere, there are many classes of scattering geometries ensuring the validity of Eq. (27) but, for conciseness, we here discuss only two main examples.

The first class we consider contains the scattering geometries, sketched in Fig. 2(a), whose outgoing polariton triads have $\mathbf{N}^2 = -\mathbf{N}^1$ and an arbitrary $\mathbf{e}_{\mathbf{N}^i\lambda}$ ($i = 1, 2$) and whose ingoing polariton triads are mutually related by a π rotation around $\mathbf{N}^i$, i.e., $\mathbf{m}^2 = \mathcal{R} \cdot \mathbf{m}^1$ and $\mathbf{e}_{\mathbf{m}^2\lambda} = \mathcal{R} \cdot \mathbf{e}_{\mathbf{m}^1\lambda}$ where

$\mathcal{R} = -\mathcal{I} + 2\mathbf{N}^i\mathbf{N}^i$ is the rotation dyadic. Such scattering geometries correspond to actual setups where the two detectors, with arbitrary polarization directions, are symmetrically aligned around the sphere, whereas the two input ports through which the ingoing s polaritons are launched are mutually related by a half-circle rotation around the direction of the detectors. The virtue of such scattering geometries is that the two outgoing polariton triads are very easily transformed by the above rotation because $\mathcal{R} \cdot \mathbf{N}^i = \mathbf{N}^i$ and $\mathcal{R} \cdot \mathbf{e}_{\mathbf{N}^i\Lambda} = -\mathbf{e}_{\mathbf{N}^i\Lambda}$ so that, from the first of Eq. (22), we readily get the relation $s_{\mathbf{m}^2\lambda}^{\mathbf{N}^i\Lambda} = -s_{\mathbf{m}^1\lambda}^{\mathbf{N}^i\Lambda}$. Accordingly, Eq. (27) holds with the $+$ sign for any wave-packet spectrum $\phi_{\lambda^1\lambda^2}^\sigma(\omega^1, \omega^2)$ and the probability density of polariton-coincidence detection in the first of Eq. (12) becomes

$$\frac{dP_{ss}^\sigma(\omega^1\mathbf{N}^1\Lambda^1, \omega^2\mathbf{N}^2\Lambda^2)}{d\omega^1 d\Omega_{\mathbf{N}^1} d\omega^2 d\Omega_{\mathbf{N}^2}} = (1 + \sigma)^2 \frac{\Delta\sigma^2}{2} \left| \sum_{\lambda^1\lambda^2} \phi_{\lambda^1\lambda^2}^\sigma(\omega^1, \omega^2) s_{\mathbf{m}^1\lambda^1}^{\mathbf{N}^1\Lambda^1} s_{\mathbf{m}^2\lambda^2}^{\mathbf{N}^2\Lambda^2} \right|^2. \quad (28)$$

We conclude that, in a scattering geometry of this class, for any kind of ingoing s polaritons correlation, the coincidence detection of two outgoing polaritons of any frequency and any polarization shows perfect constructive or destructive interference according to whether the spectrum is symmetric $\sigma = 1$ or antisymmetric $\sigma = -1$, respectively.

The second class we consider contains the scattering geometries, sketched in Fig. 2(b), whose outgoing polariton triads have arbitrary $\mathbf{N}^i$ ($i = 1, 2$), $\mathbf{e}_{\mathbf{N}^1\lambda} = \mathbf{e}_{\mathbf{N}^2\lambda}$ orthogonal to the $\mathbf{N}^1\mathbf{N}^2$ plane and $\mathbf{e}_{\mathbf{N}^2\lambda} = \mathbf{N}^2 \times \mathbf{e}_{\mathbf{N}^1\lambda}$ and whose ingoing polariton triads are mutually related by a reflection through the $\mathbf{N}^1\mathbf{N}^2$ plane, i.e., $\mathbf{m}^2 = \mathcal{W} \cdot \mathbf{m}^1$ and $\mathbf{e}_{\mathbf{m}^2\lambda} = \mathcal{W} \cdot \mathbf{e}_{\mathbf{m}^1\lambda}$ where $\mathcal{W} = \mathcal{I} - 2\mathbf{e}_{\mathbf{N}^1\lambda}\mathbf{e}_{\mathbf{N}^1\lambda}$ is the reflection dyadic. In an actual setup corresponding to a scattering geometry of this class, the two detectors can be arbitrarily placed in space with their polarization direction either belonging to or orthogonal to the detector plane, whereas the two input ports through which

the ingoing s polaritons are launched are specular images of each other with respect to the detector plane. In a scattering geometry of this class, the two outgoing polariton triads are transformed by the above reflection as $\mathcal{W} \cdot \mathbf{N}^i = \mathbf{N}^i$ and $\mathcal{W} \cdot \mathbf{e}_{\mathbf{N}^i\Lambda} = (-\delta_{\Lambda 1} + \delta_{\Lambda 2})\mathbf{e}_{\mathbf{N}^i\Lambda}$ so that the first of Eq. (22) readily implies that $s_{\mathbf{m}^i\Lambda}^{\mathbf{N}^i\Lambda} = (-\delta_{\Lambda 1} + \delta_{\Lambda 2})s_{\mathbf{m}^i\Lambda}^{\mathbf{N}^i\Lambda}$. Therefore, Eq. (27) holds with sign $(-\delta_{\Lambda 1} + \delta_{\Lambda 2})(-\delta_{\Lambda 2} + \delta_{\Lambda 1}) = 2\delta_{\Lambda 1\Lambda 2} - 1$ for any wave-packet spectrum $\phi_{\lambda^1\lambda^2}^{\sigma}(\omega^1, \omega^2)$ and the probability density of polariton-coincidence detection in the first of Eq. (12) becomes

$$\begin{aligned} & \frac{dP_{ss}^{\sigma}(\omega^1\mathbf{N}^1\Lambda^1, \omega^2\mathbf{N}^2\Lambda^2)}{d\omega^1 d\omega_{\mathbf{N}^1} d\omega^2 d\omega_{\mathbf{N}^2}} \\ &= [1 + \sigma(2\delta_{\Lambda^1\Lambda^2} - 1)]^2 \frac{\Delta\sigma^2}{2} \left| \sum_{\lambda^1\lambda^2} \phi_{\lambda^1\lambda^2}^{\sigma}(\omega^1, \omega^2) s_{\mathbf{m}^1\Lambda^1}^{\mathbf{N}^1\Lambda^1} s_{\mathbf{m}^2\Lambda^2}^{\mathbf{N}^2\Lambda^2} \right|^2. \end{aligned} \quad (29)$$

In plain English, in a scattering geometry of this class, for any kind of ingoing s polaritons correlation, the coincidence detection of two outgoing polaritons of any frequency and with the same polarization type $\Lambda^1 = \Lambda^2$ shows perfect constructive or destructive interference according to whether the spectrum is symmetric $\sigma = 1$ or antisymmetric $\sigma = -1$, respectively. Conversely, the coincidence detection of two outgoing polaritons of any frequency and with opposite polarization types $\Lambda^1 \neq \Lambda^2$ shows perfect constructive or destructive interference according to whether the spectrum is antisymmetric $\sigma = -1$ or symmetric $\sigma = 1$, respectively.

It is worth comparing such results with two-photon interference in a 50-50 lossless beam splitter [17] which is well known to yield perfect destructive interference or Hong-Ou-Mandel effect (photon coalescence) and perfect constructive interference (photon anticoalescence) according to whether the ingoing two-photon state is symmetric or antisymmetric, respectively. A first striking difference is that the key equality of the transmission and reflection rates is the only symmetry requirement enabling the beam splitter to yield perfect interference, whereas the sphere, due to its uncountable number of input-output ports and higher symmetry, dramatically admits a plenty of suitable scattering geometries, as discussed above. Second, the beam splitter transparency requirement uniquely links perfect constructive and destructive interference to symmetric and antisymmetric ingoing two-photon states, respectively, whereas, in the case of the sphere, the relation between perfect interference type and ingoing spectrum symmetry generally depends on the selected scattering geometry and the chosen detection polarizations.

As a final remark, we stress that perfect interference in the polariton-coincidence detection is evidently an idealization since, in real setups, a spherical sample always displays slight geometrical deviations from the ideal case so that its permittivity is $\varepsilon_{\omega}(\mathbf{s}) = [H(s-a) + \varepsilon_{\omega}^V H(a-s)] + \delta\varepsilon_{\omega}(\mathbf{s})$ where H is the Heaviside step function and $\delta\varepsilon_{\omega}(\mathbf{s})$ is a first-order perturbation to the rotational invariant zeroth-order permittivity, i.e., $|\delta\varepsilon_{\omega}(\mathbf{s})| \ll |\varepsilon_{\omega}^V|$. However, due to the linearity of classical

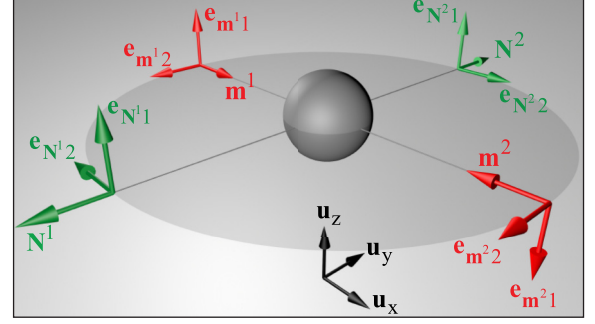


FIG. 3. Selected scattering geometry for discussing the spectral features of quantum interference effects. The ingoing polariton triads $\{\mathbf{m}^i, \mathbf{e}_{\mathbf{m}^i}, \mathbf{e}_{\mathbf{m}^i}\}$, defining the input ports, are plotted in red, whereas the outgoing polariton triads $\{\mathbf{N}^i, \mathbf{e}_{\mathbf{N}^i}, \mathbf{e}_{\mathbf{N}^i}\}$, indicating the detection ports, are depicted in green. The black arrows represent the unit vectors of the Cartesian axes.

electrodynamics, the perturbation $\delta\varepsilon_{\omega}(\mathbf{s})$ produces first-order corrections $\delta s_{\mathbf{n}\Lambda}^{\mathbf{N}\Lambda}(\omega)$ to the scattering coefficients of Eq. (9) so that Eq. (27) is satisfied only at order zero. Bearing in mind that the perfect interference condition is nonresonant in nature, i.e., Eq. (27) does not result from the vanishing of some denominators, we conclude that geometric deviations from the ideal case will slightly affect the peaks/dips signaling perfect constructive/destructive interference and showing up in the polariton-coincidence detection upon continuous sweep of the geometrical parameters. It should be stressed that, for 50/50 lossless beam splitters, an analogous phenomenology accompanies the detection of perfect destructive interference, which, owing to the inevitable deviations from the ideal case, is well known to be signaled by notable Hong-Ou-Mandel dips.

VI. SPECTRAL FEATURES OF QUANTUM INTERFERENCE EFFECTS

The major virtue of the quantum optical scattering approach (and of the MLNF) reviewed in Sec. II is that it enables to investigate the impact of macroscopic matter dispersion/absorption on quantum phenomena. Accordingly, we here model the sphere permittivity by means of the Drude-Lorentz model $\varepsilon_{\omega}^V = 1 + \omega_p^2/(\omega_0^2 - \omega^2 - i\omega\gamma)$, and we analyze the impact of the sphere dispersion on the quantum interference effects pertaining to polariton-coincidence detection and global scattering processes. To both reduce the analysis complexity and to highlight the interference effects, we select the specific scattering geometry, sketched in Fig. 3, whose ingoing and outgoing polariton triads are $\mathbf{m}^i = (\delta_{i1} - \delta_{i2})\mathbf{u}_x$, $\mathbf{e}_{\mathbf{m}^i\Lambda} = (\delta_{i1} - \delta_{i2})\delta_{\Lambda 1}\mathbf{u}_z - \delta_{\Lambda 2}\mathbf{u}_y$ and $\mathbf{N}^i = (-\delta_{i1} + \delta_{i2})\mathbf{u}_y$, $\mathbf{e}_{\mathbf{N}^i\Lambda} = \delta_{\Lambda 1}\mathbf{u}_z + (-\delta_{i1} + \delta_{i2})\delta_{\Lambda 2}\mathbf{u}_x$.

By using the Legendre polynomials properties $P_n'(0) = nP_{n-1}(0)$, $P_n''(0) = -n(n+1)P_n(0)$, and $P_{2m+1}(0) = 0$, the scattering coefficient in the first of Eq. (22) for such a scattering geometry turns out to be

$$s_{\mathbf{m}^i\Lambda}^{\mathbf{N}^i\Lambda}(\omega) = (-\delta_{j1} + \delta_{j2})(\delta_{\Lambda 1}\delta_{\Lambda 1}T_{\omega}^{oe} + \delta_{\Lambda 2}\delta_{\Lambda 2}T_{\omega}^{eo}), \quad (30)$$

where

$$\begin{aligned}
 T_{\omega}^{oe} &= \frac{1}{2\pi} \sum_{m=0}^{\infty} \left(\frac{4m+3}{2m+2} \right) P_{2m}(0) a_{(2m+1)\omega} \\
 &\quad + \frac{1}{2\pi} \sum_{m=1}^{\infty} (4m+1) P_{2m}(0) b_{(2m)\omega}, \\
 T_{\omega}^{eo} &= \frac{1}{2\pi} \sum_{m=1}^{\infty} (4m+1) P_{2m}(0) a_{(2m)\omega} \\
 &\quad + \frac{1}{2\pi} \sum_{m=0}^{\infty} \left(\frac{4m+3}{2m+2} \right) P_{2m}(0) b_{(2m+1)\omega}, \quad (31)
 \end{aligned}$$

whose superscripts emphasize their dependencies on the Mie coefficients, i.e., T_{ω}^{oe} depends on the odd $a_{n\omega}$ and the even $b_{n\omega}$, whereas T_{ω}^{eo} depends on the even $a_{n\omega}$ and the odd $b_{n\omega}$. In turn, the probability density of polariton-coincidence detection in the first of Eq. (12) becomes

$$\begin{aligned}
 &\frac{dP_{ss}^{\sigma}(\omega^1 \mathbf{N}^1 \Lambda^1, \omega^2 \mathbf{N}^2 \Lambda^2)}{d\omega^1 d\mathbf{N}^1 d\omega^2 d\mathbf{N}^2} \\
 &= (1 + \sigma)^2 \frac{\Delta\sigma^2}{2} \begin{vmatrix} \delta_{\Lambda^1 1} & \delta_{\Lambda^1 2} \end{vmatrix} \\
 &\quad \times \begin{pmatrix} \phi_{11}^{\sigma}(\omega^1, \omega^2) (T_{\omega}^{oe})^2 & \phi_{12}^{\sigma}(\omega^1, \omega^2) T_{\omega}^{oe} T_{\omega}^{eo} \\ \phi_{21}^{\sigma}(\omega^1, \omega^2) T_{\omega}^{oe} T_{\omega}^{eo} & \phi_{22}^{\sigma}(\omega^1, \omega^2) (T_{\omega}^{eo})^2 \end{pmatrix} \begin{pmatrix} \delta_{\Lambda^2 1} \\ \delta_{\Lambda^2 2} \end{pmatrix}, \quad (32)
 \end{aligned}$$

which evidently vanishes for $\sigma = -1$ since the here considered scattering geometry of Fig. 3 belongs to the first class discussed in Sec. IV. Note that such a probability density for each polarization pair (Λ^1, Λ^2) is proportional to the squared modulus of the corresponding entry in the inner matrix.

By using the Legendre polynomials property $P'_n(\pm 1) = (\pm 1)^{n+1} n(n+1)/2$, the power scattering and absorption coefficients in the second and third of Eq. (22) for the considered scattering geometry become

$$\begin{aligned}
 S_{\mathbf{m}'\lambda'}^{\mathbf{m}\lambda}(\omega) &= \delta_{ij} \delta_{\lambda\lambda'} L_{\omega}^{+} + (1 - \delta_{ij})(-\delta_{\lambda 1} \delta_{\lambda' 1} + \delta_{\lambda 2} \delta_{\lambda' 2}) L_{\omega}^{-}, \\
 A_{\mathbf{m}'\lambda'}^{\mathbf{m}\lambda}(\omega) &= \delta_{ij} \delta_{\lambda\lambda'} K_{\omega}^{+} + (1 - \delta_{ij})(-\delta_{\lambda 1} \delta_{\lambda' 1} + \delta_{\lambda 2} \delta_{\lambda' 2}) K_{\omega}^{-}, \quad (33)
 \end{aligned}$$

where

$$\begin{aligned}
 L_{\omega}^{\pm} &= \frac{1}{2\pi} \sum_{n=1}^{\infty} (\pm 1)^{n+1} (2n+1) (|a_{n\omega}|^2 \pm |b_{n\omega}|^2), \\
 K_{\omega}^{\pm} &= \frac{1}{2\pi} \sum_{n=1}^{\infty} (\pm 1)^{n+1} (2n+1) \{ [\text{Re}(a_{n\omega}) \\
 &\quad - |a_{n\omega}|^2] \pm [\text{Re}(b_{n\omega}) - |b_{n\omega}|^2] \}, \quad (34)
 \end{aligned}$$

quantities whose physical meaning is directly provided by the discussion in Sec. III C. Specifically, from Eq. (33), L_{ω}^{+} and K_{ω}^{+} are the diagonal terms of the power scattering and absorption coefficients, and they represent the classical scattering and absorption cross sections of a single plane wave, in agreement with Eq. (26), whereas L_{ω}^{-} and K_{ω}^{-} are the

off-diagonal terms representing the classical interference of two plane waves in the considered scattering geometry. By exploiting the spectral overlap integral properties in Eq. (17), the polariton scattering probabilities of Eq. (14) become

$$\begin{aligned}
 P_{2s}^{\sigma} &= \Delta\sigma^2 \{ (L_{\omega}^{+})^2 + \sigma [1 - 4(I^{\sigma})_{12}^{12}] (L_{\omega}^{-})^2 \}, \\
 P_{1s}^{\sigma} &= 2\Delta\sigma^2 \{ L_{\omega}^{+} K_{\omega}^{+} + \sigma [1 - 4(I^{\sigma})_{12}^{12}] L_{\omega}^{-} K_{\omega}^{-} \}, \\
 P_{0s}^{\sigma} &= \Delta\sigma^2 \{ (K_{\omega}^{+})^2 + \sigma [1 - 4(I^{\sigma})_{12}^{12}] (K_{\omega}^{-})^2 \}, \quad (35)
 \end{aligned}$$

whose structure, in view of the physical meaning of $L_{\omega}^{\pm}$ and $K_{\omega}^{\pm}$, unveils the role played by classical plane waves' interference in the emergence of quantum interference effects. In fact, in each of the scattering probabilities of Eq. (35), the noninterference (first) contributions contain the correct number of single plane-wave scattering and absorption cross sections L_{ω}^{+} and K_{ω}^{+} whereas the interference (second) contributions, with the factor σ , contain the correct number of classical two plane-wave interference terms L_{ω}^{-} and K_{ω}^{-} . Note that the scattering probabilities of Eq. (35) explicitly depend on the ingoing wave packet spectrum only through the overlap integral $(I^{\sigma})_{12}^{12} = \int d\omega^1 d\omega^2 |\phi_{12}^{\sigma}(\omega^1, \omega^2)|^2$ appearing in their interference contributions.

To explicitly discuss the spectral features of the polariton coincidence-detection probability density in Eq. (32) and of the polariton scattering probabilities of Eq. (35), we consider a sphere of radius $a = 1 \mu\text{m}$ whose Drude-Lorentz permittivity ε_{ω}^V , for wavelengths ranging from 3 to 20 μm , has its real and imaginary parts plotted in Fig. 4(a). Accordingly, the subwavelength-sized sphere is weakly dispersive with marked dielectric behavior (high and positive permittivity real part) and moderate but non-negligible losses. The purple and orange dashed vertical lines locate the two specific wavelengths $\lambda_1 = 12.6 \mu\text{m}$ and $\lambda_2 = 18.1 \mu\text{m}$, respectively, which we exploit in the following spectral analysis.

In Fig. 4(b), we report for $n = 1, 2, 3$ the plots of the square moduli $|a_{n\omega}|^2$, $|b_{n\omega}|^2$ and of the combinations $\text{Re}(a_{n\omega}) - |a_{n\omega}|^2$, $\text{Re}(b_{n\omega}) - |b_{n\omega}|^2$ of the Mie coefficients, the latter two assessing the overall impact of sphere losses on the scattering process, as discussed in Sec. III D. The first striking feature of Fig. 4(b) is that the quantities of order $n = 3$ (green) are generally much smaller than those of order $n = 1$ (black) and $n = 2$ (red), so that all the higher-order ones ($n \geq 4$, not plotted here) can be neglected in the present analysis. Moreover, Fig. 4(b) shows that all the quantities globally decrease at larger wavelengths (where the sphere is electromagnetically smaller) and, most importantly, that they exhibit various Mie resonance peaks whose spectral spacing increases at larger wavelengths. It is crucial to stress here that, due to the sphere's high-index dielectric behavior, Mie resonance peaks of $|a_{n\omega}|^2$ and $|b_{n\omega}|^2$ exhibit characteristic Fano lineshapes [77] with different asymmetry degrees (tuned by the Fano parameter of the specific peak) and possibly displaying strong lateral dips where $|a_{n\omega}|^2$ and $|b_{n\omega}|^2$ drop down by several orders of magnitude (the dips reach zero only for ideally transparent spheres). Besides, Mie resonances are also accompanied by strong energy dissipation in the presence of matter losses, as evinced by the corresponding peaks in $\text{Re}(a_{n\omega}) - |a_{n\omega}|^2$ and $\text{Re}(b_{n\omega}) - |b_{n\omega}|^2$ whose spectral profiles are practically symmetric since the Fano mechanism is inhibited in the

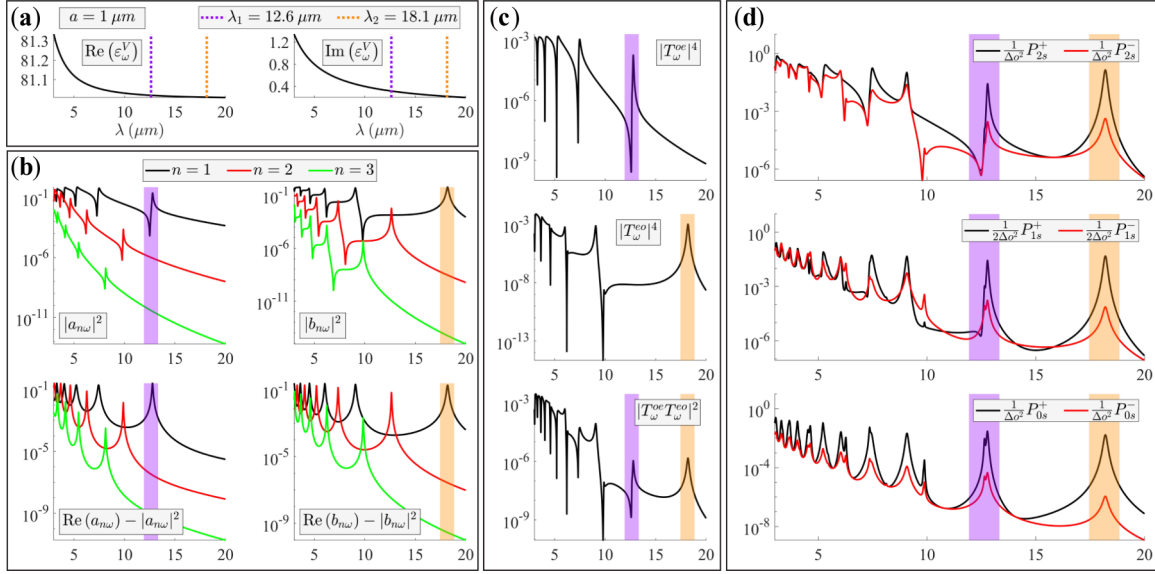


FIG. 4. Quantum optical scattering by a subwavelength-sized lossy sphere with high-index dielectric behavior. (a) Real and imaginary parts of the dielectric permittivity. (b) Square moduli $|a_{nw}|^2$, $|b_{nw}|^2$, and combinations $\text{Re}(a_{nw}) - |a_{nw}|^2$, $\text{Re}(b_{nw}) - |b_{nw}|^2$ of the Mie coefficients. (c) Plots of the quantities $|T_\omega^{oe}|^4$, $|T_\omega^{eo}|^4$, and $|T_\omega^{oe} T_\omega^{eo}|^2$ appearing in Eq. (32). Such quantities are proportional to the probability densities of coincidence detection of two polaritons with specific polarizations and, accordingly, they quantify the corresponding interference visibility. For example, for $\Lambda_1 = 1$ and $\Lambda_2 = 1$, we have $dP_{ss}^+(\omega^1 \mathbf{N}^1, \omega^2 \mathbf{N}^2)/d\omega^1 d\omega^2 d\mathbf{O}_{N^1} d\mathbf{O}_{N^2} = 2\Delta\sigma^2 |\phi_{11}^\sigma(\omega^1, \omega^2)|^2 |T_\omega^{oe}|^4$. (d) Plots of the normalized polariton scattering probabilities of Eq. (35) for ingoing photon wave packets with no spectral overlap, i.e., $(I^\sigma)_{12}^2 = 0$.

absorption cross section. Note that the wavelengths λ_1 and λ_2 introduced in Fig. 4(a) correspond to specific Mie resonances of $|a_{1\omega}|^2$ and $|b_{1\omega}|^2$, and we have highlighted the spectral regions surrounding such wavelengths by means of purple and orange-shaded rectangles, respectively.

In Fig. 4(c), we plot the combinations T_ω^{oe} and T_ω^{eo} of Eq. (31) appearing in the probability density of polariton-coincidence detection of Eq. (32), which, we repeat, vanishes for antisymmetric wave packets ($\sigma = -1$) due to perfect destructive interference, so that we here focus on symmetric wave packets ($\sigma = +1$). For example, $|T_\omega^{oe}|^4$ basically lends its spectral features to the probability density of coincide detection of the outgoing polaritons of directions $\mathbf{N}^1 = -\mathbf{u}_y$, $\mathbf{N}^2 = \mathbf{u}_y$ and polarizations $\mathbf{e}_{N^1} = \mathbf{e}_{N^2} = \mathbf{u}_z$ (see Fig. 3). The most important result shown by Fig. 4(c) is that the Mie resonances yield marked spectral peaks in the probability density of coincidence detection of any pair of outgoing polaritons, which physically means that classical resonances are responsible for large quantum constructive interference. The spectral features of such peaks can easily be grasped after noting that, for the Mie coefficients in Fig. 4(b), Eq. (31) can be approximated as $T_\omega^{oe} \simeq \frac{3}{4\pi} P_0(0) a_{1\omega}$ and $T_\omega^{eo} \simeq \frac{3}{4\pi} P_0(0) b_{1\omega}$ so that, for example, the peak at λ_1 (purple) of $|T_\omega^{oe}|^4$ and at λ_2 (orange) of $|T_\omega^{eo}|^4$ are produced by the corresponding Mie resonances of $a_{1\omega}$ and $b_{1\omega}$, respectively (and such two peaks evidently show up together in $|T_\omega^{oe} T_\omega^{eo}|^2$). It is also worth noting that the possible asymmetric line-shapes of the Mie peaks in $|a_{nw}|^2$ and $|b_{nw}|^2$, physically due to the Fano mechanism, correspondingly yields significant dips in Fig. 4(c), representing spectral regions where very large quantum destructive interference occurs, as the one in $|T_\omega^{oe}|^4$, which is close to the resonance wavelength λ_1 (purple)

whose minimum is about six orders of magnitude smaller than the nearby maximum reached at resonance. It is also remarkable that the almost symmetric peak in $|T_\omega^{eo}|^4$ at the wavelength λ_2 (orange) has a maximum which is about five orders of magnitude larger than its neighbor off-resonance background.

In Fig. 4(d), we analyze the polariton scattering probabilities of Eq. (35), for ingoing wave packets with no spectral overlap, i.e., $(I^\sigma)_{12}^2 = 0$ (for the sake of simplicity), by plotting the quantities $(L_\omega^+)^2 + \sigma(L_\omega^-)^2$, $L_\omega^+ K_\omega^+ + \sigma L_\omega^- K_\omega^-$, and $(K_\omega^+)^2 + \sigma(K_\omega^-)^2$, using black and red solid lines for symmetric $\sigma = +1$ and antisymmetric $\sigma = -1$ wave packets, respectively. Evidently, in close analogy with the polariton-coincidence detection, also the polariton scattering probabilities exhibit marked peaks associated with the classical Mie resonances, and such peaks can display lateral deep dips due to the Fano mechanism accompanying $|a_{nw}|^2$ and $|b_{nw}|^2$. In other words, classical Mie resonances have a large impact on quantum constructive and destructive interference effects accompanying the global polariton scattering processes. However, the results reported in Fig. 4(d) are also interesting for two main additional reasons. First, the one and zero polariton scattering probabilities P_{1s}^σ and P_{0s}^σ are strictly vanishing for an ideally transparent sphere (and for any transparent object), so that their spectral features provide additional routes to perform quantum measurements which are not available when matter losses can be neglected. Second, Fig. 4(d) makes it clear that the polariton scattering probabilities for symmetric (black lines) and antisymmetric (red lines) wave packets are generally different, their discrepancy being very large at the resonance peaks of a single Mie coefficient, as the peaks within the orange shaded rectangles. Such large discrepancies

occur since, close to resonances of this kind, the polariton survival probabilities of Eq. (35) for antisymmetric wave packets $\sigma = -1$ are very small due to the physical fact that the interference terms $|L_\omega^-|$ and $|K_\omega^-|$ nearly equate the scattering and absorption cross sections $|L_\omega^+|$ and $|K_\omega^+|$, respectively. As an example, the first of Eq. (34) for the Mie coefficients in Fig. 4(b) yields $|L_\omega^\pm| \simeq \frac{3}{2\pi} |a_{1\omega}|^2 \pm |b_{1\omega}|^2$ so that, close to the wavelength λ_2 where $|b_{1\omega}|^2 \gg |a_{1\omega}|^2$ (i.e., only $b_{1\omega}$ resonates), we have that $|L_\omega^+| \simeq |L_\omega^-|$ and most importantly that

$$\frac{(L_\omega^+)^2 - (L_\omega^-)^2}{(L_\omega^+)^2 + (L_\omega^-)^2} \simeq 2 \frac{|a_{1\omega}|^2}{|b_{1\omega}|^2} \ll 1, \quad (36)$$

thus explaining the three orders of magnitude discrepancy between the (red and black) peak values at the wavelength λ_2 (orange shaded rectangle) in the first subplot of Fig. 4(d). We conclude that the quantum interference effects in the global polariton scattering processes produced by classical Mie resonances exhibit marked sensitivity to the symmetry of the wave packet spectrum, a characteristic that could be exploited in an actual setup to efficiently detect polariton entanglement.

VII. DISCUSSION

A final discussion on the validity of the narrow bandwidth approximation used in the present paper is here in order since, in this section, we have considered Mie resonance peaks generally exhibiting very sharp spectral features so that, in principle, wave packets with sufficiently narrow spectra could not be available. While this is a very delicate issue when resonance peaks are due to atomic or molecular transitions, quasimonochromatic pulses able to resolve the resonance line shape associated with photonic modes of macroscopic structures are available (see Refs. [82–86]) mainly for two reasons: First, the spectral features of modal resonances can be steered by tailoring the geometry and composition of the photonic structure and, second, the existence of material losses (which is among the main interests of the present paper) has the general effect of broadening the resonance line shape, thus making it easier to justify and to exploit the narrow bandwidth approximation.

Regarding a possible experimental implementation of the quantum interference phenomena described thus far, it is instructive to discuss the feasibility of the theoretical assumptions underlying our model. First, the generation of wave packets with definite spectral symmetry ($\sigma = \pm 1$) is a well-established procedure in modern quantum optics. Spectrally symmetric or antisymmetric two-photon states are routinely employed in standard Hong-Ou-Mandel experiments with classical beam splitters, and they can be obtained by tailoring the biphoton wave function produced via spontaneous parametric down-conversion (SPDC) or through coherent spectral pulse-shaping techniques. Regarding the scattering geometries required to observe perfect interference in the polariton-coincidence detection, standard optical alignment protocols can be effectively adopted. The use of piezoelectric nanopositioning stages coupled with confocal microscopy setups allows for centering the single nanoparticle relative to the input and output channels with subwavelength precision,

thus ensuring the interferometric stability demanded by the parity or rotation transformations. Furthermore, although the spectral features of the quantum interference effects have been discussed here for the regular geometry of Fig. 3 to provide immediate physical intuition and avoid overburdening the mathematical treatment, the underlying physical mechanism is completely general. The significant discrepancy between the scattering probabilities associated with symmetric and antisymmetric impinging wave packets persists even in scattering configurations lacking specific spatial symmetries. The crucial condition of well-separated and isolated Mie resonances is experimentally accessible using high-index dielectric spheres, such as silicon or germanium. A wide variety of such materials exists, exhibiting diverse optical characteristics: some are intrinsically dissipative, while others are almost perfectly transparent in specific frequency ranges. This variety allows for a precise mapping of the discrepancy between the lossy and the ideal lossless regimes. It is fundamental to emphasize that the simultaneous detection of two photons, only a single photon, or exactly zero photons (corresponding to the *ss*, *se*, and *ee* channels, respectively) are among the fundamental measurements routinely performed in quantum optics. Moreover, the specific idea of independently measuring these three distinct channels to characterize dissipative quantum interference has already been proposed and investigated for the case of the lossy four-port device (see Ref. [36]). In the present work, we repurpose this measurement strategy for the case of a realistic sphere, which, as we have demonstrated, exhibits physical behaviors that are completely different from those characterizing the standard four-port device. Ultimately, the primary experimental goal lies in measuring the macroscopic signature of this quantum interference effect, which manifests as a massive difference in coincidence probabilities between symmetric and antisymmetric impinging wave packets. Importantly, the experimental discrimination of these two cases is not hindered by detector sensitivity limitations. Because the corresponding scattering probabilities are predicted to differ by several orders of magnitude, this stark contrast guarantees a high signal-to-noise ratio, ensuring that the effect remains unambiguously resolvable even against typical experimental background noise.

VIII. CONCLUSION

In conclusion, we have exploited a recently introduced quantum optical scattering framework, based on the MLNF, to analyze the scattering of two directional *s* polaritons by a macroscopic lossy sphere, which, as any other finite-size scatterer, can effectively be regarded as an optical device with an infinite number of input and output ports associated with the ingoing polariton directions. Classical matter losses physically yield the quantum mechanism of *s* polariton absorption, thus enabling the existence of three distinct processes where two, one, or zero *s* polaritons survive the scattering. If the ingoing *s* polaritons are not collinear, the detections of such three processes display marked quantum interference effects which represent a far-reaching generalization of those supported by standard beam splitters since their efficient observation is facilitated by the much greater freedom of choosing the scattering geometry (i.e., the input

and output ports). As an example, the equality of transmission and reflection rates is the only condition enabling the beam splitter to exhibit the famous Hong-Ou-Mandel effect, whereas we have here proved that different classes of scattering geometries allow the sphere to perform perfect destructive interference. Exploiting the MLNF ability to account for macroscopic matter dispersive response, we have discussed the impact of the sphere losses on the quantum interference effects arising in the detection of the scattered radiation. In particular, we have shown that classical Mie resonances dramatically provide spectral bandwidths where very large constructive and destructive interference occurs. In analogy to the above-discussed flexibility of the geometric setup provided by the sphere, this result is also a noteworthy generalization of the spectral abilities of a lossy beam splitter since it usually does not exhibit resonances and it has only been shown to range from photon coalescence to anticoincidence. As a final remark, we stress that matter losses open up the possibility of performing measurements not allowed by ideally transparent objects, such as, for instance, the overall detection of one or zero scattered s polariton in the due polaritons scattering considered in this paper. As previously established for lossy beam splitters [38–40], material loss in the quantum regime does not merely cause a passive statistical degradation of the signal, but physically opens up new, genuine quantum channels (involving the excitation of matter polaritons). We have shown that the corresponding scattering probabilities are very sensitive to the spectral symmetry of the ingoing wave packet when its central frequency matches an isolated Mie resonance. Since, in this case, the discrepancy between the scattering probabilities of symmetric and anti-symmetric wave packets can be several orders of magnitude, this phenomenology provides a specific application: using macroscopic material loss as an active spectral filter to efficiently detect polariton entanglement simply by monitoring the single-photon or zero-photon survival rates.

DATA AVAILABILITY

There are no publicly available research data or software supporting this paper. Requests for further information or data should be sent to the authors.

APPENDIX A: MODIFIED LANGEVIN NOISE FORMALISM AND QUANTUM OPTICAL SCATTERING APPROACH

As detailed in Ref. [73], the basic tools MLNF borrows from classical electrodynamics are the modal dyadic $\mathcal{F}_\omega(\mathbf{s}|\mathbf{n})$ and the dyadic Green's function $\mathcal{G}_\omega(\mathbf{s}|\mathbf{r})$, which are defined by the boundary-value problems

$$\begin{aligned} [(\nabla_s \times \nabla_s \times) - k_\omega^2 \varepsilon_\omega(\mathbf{s})] \mathcal{F}_\omega(\mathbf{s}|\mathbf{n}) &= 0, \\ \mathcal{F}_\omega(\mathbf{s}|\mathbf{n}) &\underset{s \rightarrow +\infty}{\approx} e^{i(k_\omega s) \mathbf{n} \cdot \mathbf{n}} \mathcal{I}_\mathbf{n} + \frac{e^{ik_\omega s}}{s} \mathcal{S}_\omega(\mathbf{N}|\mathbf{n}), \\ [(\nabla_s \times \nabla_s \times) - k_\omega^2 \varepsilon_\omega(\mathbf{s})] \mathcal{G}_\omega(\mathbf{s}|\mathbf{r}) &= \delta(\mathbf{s} - \mathbf{r}) \mathcal{I}, \\ \mathcal{G}_\omega(\mathbf{s}|\mathbf{r}) &\underset{s \rightarrow +\infty}{\approx} \frac{e^{ik_\omega s}}{s} \mathcal{W}_\omega(\mathbf{N}|\mathbf{r}), \end{aligned} \quad (\text{A1})$$

where $k_\omega = \omega/c$ is the vacuum wave number, $\mathbf{N}$ and $\mathbf{n}$ are unit vectors, $\mathcal{I}$ is the identity dyadic, $\mathcal{I}_\mathbf{n} = \mathcal{I} - \mathbf{n}\mathbf{n}$ is the dyadic projector onto the plane orthogonal to $\mathbf{n}$, and the symbol $s \underset{s \rightarrow +\infty}{\approx}$ stands for the leading-order term of the asymptotic expansion for $s \rightarrow +\infty$. We hereafter denote with $do_\mathbf{n} = \sin \theta_\mathbf{n} d\theta_\mathbf{n} d\varphi_\mathbf{n}$ the differential of the solid angle around the unit vector $\mathbf{n} = \sin \theta_\mathbf{n} (\cos \varphi_\mathbf{n} \mathbf{u}_x + \sin \varphi_\mathbf{n} \mathbf{u}_y) + \cos \theta_\mathbf{n} \mathbf{u}_z$. The modal dyadic satisfies the orthogonality relation $\mathcal{F}_\omega(\mathbf{s}|\mathbf{n}) \cdot \mathbf{n} = 0$, and the dyadic $\mathcal{S}_\omega(\mathbf{N}|\mathbf{n})$, appearing in its asymptotic behavior, is the classical scattering dyadic [80] satisfying the orthogonality relations $\mathbf{N} \cdot \mathcal{S}_\omega(\mathbf{N}|\mathbf{n}) = 0$ and $\mathcal{S}_\omega(\mathbf{N}|\mathbf{n}) \cdot \mathbf{n} = 0$. The dyadic $\mathcal{W}_\omega(\mathbf{N}|\mathbf{r})$ is the asymptotic amplitude of the dyadic Green's function satisfying the orthogonal relation $\mathbf{N} \cdot \mathcal{W}_\omega(\mathbf{N}|\mathbf{r}) = 0$, and it is related to the modal dyadic by the remarkable relation $\mathcal{W}_\omega(\mathbf{N}|\mathbf{r}) = \frac{1}{4\pi} \mathcal{F}_\omega^T(\mathbf{r}|\mathbf{N})$, which has been proved in Ref. [73] and is used there to analytically substantiate the MLNF. The modal dyadic and the dyadic Green's function provide the description of any classical scattering and radiation process in the presence of the object since, if $\mathbf{E}_\omega^{(\text{in})}(\mathbf{s}) = \int do_\mathbf{n} e^{i(k_\omega \mathbf{n}) \cdot \mathbf{s}} \tilde{\mathbf{E}}_\omega^{(\text{in})}(\mathbf{n})$ (with $\mathbf{n} \cdot \tilde{\mathbf{E}}_\omega^{(\text{in})}(\mathbf{n}) = 0$) is any free field impinging onto the object and $\mathbf{J}_\omega(\mathbf{s})$ is the current density of any radiation source, the overall field throughout space is

$$\begin{aligned} \mathbf{E}_\omega(\mathbf{s}) &= \int do_\mathbf{n} \mathcal{F}_\omega(\mathbf{s}|\mathbf{n}) \cdot \tilde{\mathbf{E}}_\omega^{(\text{in})}(\mathbf{n}) \\ &\quad + i\omega\mu_0 \int d^3\mathbf{r} \mathcal{G}_\omega(\mathbf{s}|\mathbf{r}) \cdot \mathbf{J}_\omega(\mathbf{r}), \end{aligned} \quad (\text{A2})$$

i.e., the superposition of the scattered (first contribution) and radiated (second contribution) fields. From the right column of Eq. (A1), the asymptotic behavior of such a field is $\mathbf{E}_\omega(\mathbf{s}|\mathbf{N}) \underset{s \rightarrow +\infty}{\approx} \mathbf{E}_\omega^{(\text{in})}(\mathbf{s}|\mathbf{N}) + \frac{e^{ik_\omega s}}{s} [\mathbf{f}_\omega^{\text{sca}}(\mathbf{N}) + \mathbf{f}_\omega^{\text{rad}}(\mathbf{N})]$ where

$$\begin{aligned} \mathbf{f}_\omega^{\text{sca}}(\mathbf{N}) &= \int do_\mathbf{n} \mathcal{S}_\omega(\mathbf{N}|\mathbf{n}) \cdot \tilde{\mathbf{E}}_\omega^{(\text{in})}(\mathbf{n}), \\ \mathbf{f}_\omega^{\text{rad}}(\mathbf{N}) &= i\omega\mu_0 \int d^3\mathbf{r} \mathcal{W}_\omega(\mathbf{N}|\mathbf{r}) \cdot \mathbf{J}_\omega(\mathbf{r}) \end{aligned} \quad (\text{A3})$$

are its far-field scattering and radiation amplitudes.

We hereafter denote with $\mathbf{u}_v$ the three ($v = 1, 2, 3$) Cartesian unit vectors spanning the space ($\sum_v \mathbf{u}_v \mathbf{u}_v = \mathcal{I}$) and with $\mathbf{e}_{\mathbf{n}\lambda}$ two ($\lambda = 1, 2$) orthonormal real unit vectors orthogonal to $\mathbf{n}$ ($\mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{e}_{\mathbf{n}\lambda'} = \delta_{\lambda\lambda'}$, $\mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{n} = 0$) and hence spanning the plane orthogonal to $\mathbf{n}$ ($\sum_\lambda \mathbf{e}_{\mathbf{n}\lambda} \mathbf{e}_{\mathbf{n}\lambda} = \mathcal{I}_\mathbf{n}$). Such unit vectors enable the introduction of the classical fields

$$\begin{aligned} \mathbf{F}_s(\mathbf{s}|\omega\mathbf{n}\lambda) &= \sqrt{\frac{\hbar k_\omega^3}{16\pi^3 \varepsilon_0}} \mathcal{F}_\omega(\mathbf{s}|\mathbf{n}) \cdot \mathbf{e}_{\mathbf{n}\lambda}, \\ \mathbf{F}_e(\mathbf{s}|\omega\mathbf{r}v) &= i\sqrt{\frac{\hbar k_\omega^4}{\pi \varepsilon_0} \text{Im}[\varepsilon_\omega(\mathbf{r})]} \mathcal{G}_\omega(\mathbf{s}|\mathbf{r}) \cdot \mathbf{u}_v \end{aligned} \quad (\text{A4})$$

in such a way that, apart from the numerical coefficients introduced for later quantization purposes, $\mathbf{F}_s(\mathbf{s}|\omega\mathbf{n}\lambda)$ is the field excitation at the point $\mathbf{s}$ resulting from the scattering by the object of an incident vacuum plane wave of frequency ω , direction $\mathbf{n}$, and polarization $\mathbf{e}_{\mathbf{n}\lambda}$, whereas $\mathbf{F}_e(\mathbf{s}|\omega\mathbf{r}v)$ is the field excitation at the point $\mathbf{s}$ produced by an electric dipole of frequency ω , located at the point $\mathbf{r}$ within the object [since $\text{Im}[\varepsilon_\omega(\mathbf{r})] = 0$ in vacuum] and direction $\mathbf{u}_v$. The

MLNF Hamiltonian operator and Heisenberg-picture electric-field operator are [73]

$$\begin{aligned}\hat{H} &= \int d\omega \hbar \omega \left[\int d\mathbf{o}_n \sum_{\lambda} \hat{a}_s^\dagger(\omega \mathbf{n} \lambda) \hat{a}_s(\omega \mathbf{n} \lambda) \right. \\ &\quad \left. + \int d^3 \mathbf{r} \sum_v \hat{a}_e^\dagger(\omega \mathbf{r} v) \hat{a}_e(\omega \mathbf{r} v) \right], \\ \hat{\mathbf{E}}(\mathbf{s}, t) &= \int d\omega e^{-i\omega t} \left[\int d\mathbf{o}_n \sum_{\lambda} \mathbf{F}_s(\mathbf{s} | \omega \mathbf{n} \lambda) \hat{a}_s(\omega \mathbf{n} \lambda) \right. \\ &\quad \left. + \int d^3 \mathbf{r} \sum_v \mathbf{F}_e(\mathbf{s} | \omega \mathbf{r} v) \hat{a}_e(\omega \mathbf{r} v) \right] + \text{H.c.}, \quad (\text{A5})\end{aligned}$$

where $\int d\omega = \int_0^{+\infty} d\omega$ denotes integration over positive frequencies, whereas $\hat{a}_s(\omega \mathbf{n} \lambda)$ and $\hat{a}_e(\omega \mathbf{r} v)$ are operators satisfying the bosonic commutation relations

$$\begin{aligned}[\hat{a}_s(\omega \mathbf{n} \lambda), \hat{a}_s^\dagger(\omega' \mathbf{n}' \lambda')] &= \delta(\omega - \omega') \delta(\mathbf{o}_n - \mathbf{o}_{n'}) \delta_{\lambda \lambda'}, \\ [\hat{a}_e(\omega \mathbf{r} v), \hat{a}_e^\dagger(\omega' \mathbf{r}' v')] &= \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}') \delta_{vv'}, \quad (\text{A6})\end{aligned}$$

where $\delta(\mathbf{o}_n - \mathbf{o}_{n'}) = \delta(\cos \theta_n - \cos \theta_{n'}) \delta(\varphi_n - \varphi_{n'})$ is the angular delta function, together with the vanishing of all the other possible commutation relations. In other words, MLNF describes the quantum optical field as an assembly of noninteracting bosons of two kinds, the s and e polaritons, which are the quanta associated with the field excitations $\mathbf{F}_s(\mathbf{s} | \omega \mathbf{n} \lambda)$ and $\mathbf{F}_e(\mathbf{s} | \omega \mathbf{r} v)$, respectively, arising out of the light-matter macroscopic coupling. Evidently, s polaritons can be regarded as the counterparts of photons in the lossy regime, whereas e polaritons can be viewed as quanta of the medium dipolar sources.

The approach to quantum optical scattering by lossy objects based on the MLNF and introduced in Ref. [75] additionally resorts to the transmission $\mathcal{T}_{\omega ss}(\mathbf{N} | \mathbf{n})$ [87], emission $\mathcal{E}_{\omega se}(\mathbf{N} | \mathbf{r})$, absorption $\mathcal{A}_{\omega es}(\mathbf{R} | \mathbf{n})$, and energy redistribution $\mathcal{Q}_{\omega ee}(\mathbf{R} | \mathbf{r})$ dyadics given by

$$\begin{aligned}\mathcal{T}_{\omega ss}(\mathbf{N} | \mathbf{n}) &= \delta(\mathbf{o}_N - \mathbf{o}_n) \mathcal{I}_n + \frac{ik_\omega}{2\pi} \mathcal{S}_\omega(\mathbf{N} | \mathbf{n}), \\ \mathcal{E}_{\omega se}(\mathbf{N} | \mathbf{r}) &= -\sqrt{4k_\omega^3 \text{Im}[\varepsilon_\omega(\mathbf{r})]} \mathcal{W}_\omega(\mathbf{N} | \mathbf{r}), \\ \mathcal{A}_{\omega es}(\mathbf{R} | \mathbf{n}) &= -\sqrt{4k_\omega^3 \text{Im}[\varepsilon_\omega(\mathbf{R})]} \mathcal{W}_\omega^T(-\mathbf{n} | \mathbf{R}), \\ \mathcal{Q}_{\omega ee}(\mathbf{R} | \mathbf{r}) &= -\delta(\mathbf{R} - \mathbf{r}) \mathcal{I} - i\sqrt{4k_\omega^4 \text{Im}[\varepsilon_\omega(\mathbf{R})] \text{Im}[\varepsilon_\omega(\mathbf{r})]} \\ &\quad \times \mathcal{G}_\omega(\mathbf{R} | \mathbf{r}), \quad (\text{A7})\end{aligned}$$

which are shown in Ref. [75] to satisfy the relations

$$\begin{aligned}\int d\mathbf{o}_N \mathcal{T}_{\omega ss}^{T*}(\mathbf{N} | \mathbf{n}) \cdot \mathcal{T}_{\omega ss}(\mathbf{N} | \mathbf{n}') &+ \int d^3 \mathbf{R} \mathcal{A}_{\omega es}^{T*}(\mathbf{R} | \mathbf{n}) \cdot \mathcal{A}_{\omega es}(\mathbf{R} | \mathbf{n}') = \delta(\mathbf{o}_n - \mathbf{o}_{n'}) \mathcal{I}_n, \\ \int d\mathbf{o}_N \mathcal{T}_{\omega ss}^{T*}(\mathbf{N} | \mathbf{n}) \cdot \mathcal{E}_{\omega se}(\mathbf{N} | \mathbf{r}) &+ \int d^3 \mathbf{R} \mathcal{A}_{\omega es}^{T*}(\mathbf{R} | \mathbf{n}) \cdot \mathcal{Q}_{\omega ee}(\mathbf{R} | \mathbf{r}) = 0,\end{aligned}$$

$$\begin{aligned}\int d\mathbf{o}_N \mathcal{E}_{\omega se}^{T*}(\mathbf{N} | \mathbf{r}) \cdot \mathcal{E}_{\omega se}(\mathbf{N} | \mathbf{r}') &+ \int d^3 \mathbf{R} \mathcal{Q}_{\omega ee}^{T*}(\mathbf{R} | \mathbf{r}) \cdot \mathcal{Q}_{\omega ee}(\mathbf{R} | \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \mathcal{I}, \quad (\text{A8})\end{aligned}$$

and

$$\begin{aligned}\int d\mathbf{o}_N \mathcal{T}_{\omega ss}(\mathbf{N} | \mathbf{n}) \cdot \mathcal{T}_{\omega ss}^{T*}(\mathbf{N}' | \mathbf{n}) &+ \int d^3 \mathbf{r} \mathcal{E}_{\omega se}(\mathbf{N} | \mathbf{r}) \cdot \mathcal{E}_{\omega se}^{T*}(\mathbf{N}' | \mathbf{r}) = \delta(\mathbf{o}_N - \mathbf{o}_{N'}) \mathcal{I}_N, \\ \int d\mathbf{o}_N \mathcal{T}_{\omega ss}(\mathbf{N} | \mathbf{n}) \cdot \mathcal{A}_{\omega es}^{T*}(\mathbf{R} | \mathbf{n}) &+ \int d^3 \mathbf{r} \mathcal{E}_{\omega se}(\mathbf{N} | \mathbf{r}) \cdot \mathcal{Q}_{\omega ee}^{T*}(\mathbf{R} | \mathbf{r}) = 0, \\ \int d\mathbf{o}_N \mathcal{A}_{\omega es}(\mathbf{R} | \mathbf{n}) \cdot \mathcal{A}_{\omega es}^{T*}(\mathbf{R}' | \mathbf{n}) &+ \int d^3 \mathbf{r} \mathcal{Q}_{\omega ee}(\mathbf{R} | \mathbf{r}) \cdot \mathcal{Q}_{\omega ee}^{T*}(\mathbf{R}' | \mathbf{r}) = \delta(\mathbf{R} - \mathbf{R}') \mathcal{I}. \quad (\text{A9})\end{aligned}$$

Equations (A8) and (A9) physically ensure, in full generality, energy conservation of the field-matter system upon classical scattering, encompassing the situations where the scatterer hosts sources able to radiate and absorb energy. In the quantum optical scattering approach, field-matter descriptions in the far past ($t \rightarrow -\infty$) and far future ($t \rightarrow +\infty$) are respectively afforded by the ingoing polariton operators $\hat{a}_s(\omega \mathbf{n} \lambda)$, $\hat{a}_e(\omega \mathbf{r} v)$ (lowercase letters), and outgoing polariton operators $\hat{A}_s(\omega \mathbf{N} \Lambda)$, $\hat{A}_e(\omega \mathbf{R} \Upsilon)$ (capital letters), the latter being linked to the former by the input-output transformation

$$\begin{aligned}\hat{A}_s(\omega \mathbf{N} \Lambda) &= \int d\mathbf{o}_n \sum_{\lambda} [\mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathcal{T}_{\omega ss}(\mathbf{N} | \mathbf{n}) \cdot \mathbf{e}_{\mathbf{n}\lambda}] \hat{a}_s(\omega \mathbf{n} \lambda) \\ &\quad + \int d^3 \mathbf{r} \sum_v [\mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathcal{E}_{\omega se}(\mathbf{N} | \mathbf{r}) \cdot \mathbf{u}_v] \hat{a}_e(\omega \mathbf{r} v), \\ \hat{A}_e(\omega \mathbf{R} \Upsilon) &= \int d\mathbf{o}_n \sum_{\lambda} [\mathbf{u}_\Upsilon \cdot \mathcal{A}_{\omega es}(\mathbf{R} | \mathbf{n}) \cdot \mathbf{e}_{\mathbf{n}\lambda}] \hat{a}_s(\omega \mathbf{n} \lambda) \\ &\quad + \int d^3 \mathbf{r} \sum_v [\mathbf{u}_\Upsilon \cdot \mathcal{Q}_{\omega ee}(\mathbf{R} | \mathbf{r}) \cdot \mathbf{u}_v] \hat{a}_e(\omega \mathbf{r} v). \quad (\text{A10})\end{aligned}$$

Note that the linear mixture of the operators $\hat{a}_s$ and $\hat{a}_e$ in the right-hand sides (RHSs) of Eq. (A10) is produced by the matrix elements of the classical dyadics of Eq. (A7). The crucial point is that Eqs. (A8) and (A9) entail that the input-output transformation in Eq. (A10) is invertible, i.e.,

$$\begin{aligned}\hat{a}_s(\omega \mathbf{n} \lambda) &= \int d\mathbf{o}_N \sum_{\Lambda} [\mathbf{e}_{\mathbf{n}\lambda} \cdot \mathcal{T}_{\omega ss}^{T*}(\mathbf{N} | \mathbf{n}) \cdot \mathbf{e}_{\mathbf{N}\Lambda}] \hat{A}_s(\omega \mathbf{N} \Lambda) \\ &\quad + \int d^3 \mathbf{R} \sum_{\Upsilon} [\mathbf{e}_{\mathbf{n}\lambda} \cdot \mathcal{A}_{\omega es}^{T*}(\mathbf{R} | \mathbf{n}) \cdot \mathbf{u}_\Upsilon] \hat{A}_e(\omega \mathbf{R} \Upsilon), \\ \hat{a}_e(\omega \mathbf{r} v) &= \int d\mathbf{o}_N \sum_{\Lambda} [\mathbf{u}_v \cdot \mathcal{E}_{\omega se}^{T*}(\mathbf{N} | \mathbf{r}) \cdot \mathbf{e}_{\mathbf{N}\Lambda}] \hat{A}_s(\omega \mathbf{N} \Lambda) \\ &\quad + \int d^3 \mathbf{R} \sum_{\Upsilon} [\mathbf{u}_v \cdot \mathcal{Q}_{\omega ee}^{T*}(\mathbf{R} | \mathbf{r}) \cdot \mathbf{u}_\Upsilon] \hat{A}_e(\omega \mathbf{R} \Upsilon) \quad (\text{A11})\end{aligned}$$

and that bosonic commutation relations are conserved, i.e.,

$$\begin{aligned} [\hat{A}_s(\omega\mathbf{N}\Lambda), \hat{A}_s^\dagger(\omega'\mathbf{N}'\Lambda')] &= \delta(\omega - \omega')\delta(\mathbf{N} - \mathbf{N}')\delta_{\Lambda\Lambda'}, \\ [\hat{A}_e(\omega\mathbf{R}\Upsilon), \hat{A}_e^\dagger(\omega'\mathbf{R}'\Upsilon')] &= \delta(\omega - \omega')\delta(\mathbf{R} - \mathbf{R}')\delta_{\Upsilon\Upsilon'}, \end{aligned} \quad (\text{A12})$$

together with the vanishing of all the other possible commutation relations. In other words, as a consequence of energy conservation upon classical scattering, the input-output transformation of Eq. (A10) is unitary and the operators $(\hat{a}_s, \hat{a}_e)$ and $(\hat{A}_s, \hat{A}_e)$ provide two different representations of the field-matter Fock space. It is worth pointing out that, as detailed in Ref. [75], the far-field asymptotics of the electric-field operator $\hat{\mathbf{E}}(\mathbf{s}, t)$ [second of Eq. (A5)] in the far past and far future are respectively given by

$$\begin{aligned} \hat{\mathbf{E}}(\xi\mathbf{n}, t) &\underset{\substack{\xi \rightarrow -\infty \\ t \rightarrow -\infty}}{\approx} \int_0^{+\infty} d\omega \sqrt{\frac{\hbar k_\omega}{4\pi\epsilon_0}} \frac{e^{ik_\omega(\xi-ct)}}{i\xi} \sum_{\Lambda} \mathbf{e}_{\mathbf{n}\Lambda} \hat{a}_s(\omega\mathbf{n}\Lambda) \\ &+ \text{H.c.}, \end{aligned} \quad (\text{A13})$$

$$\begin{aligned} \hat{\mathbf{E}}(\xi\mathbf{N}, t) &\underset{\substack{\xi \rightarrow +\infty \\ t \rightarrow +\infty}}{\approx} \int_0^{+\infty} d\omega \sqrt{\frac{\hbar k_\omega}{4\pi\epsilon_0}} \frac{e^{ik_\omega(\xi-ct)}}{i\xi} \sum_{\Lambda} \mathbf{e}_{\mathbf{N}\Lambda} \hat{A}_s(\omega\mathbf{N}\Lambda) \\ &+ \text{H.c.}, \end{aligned} \quad (\text{A14})$$

and their respective identical dependencies on $\hat{a}_s$ and $\hat{A}_s$ manifestly prove the above statement that the ingoing operators $(\hat{a}_s, \hat{a}_e)$ and the outgoing operators $(\hat{A}_s, \hat{A}_e)$ respectively provide the input and output field-matter descriptions in quantum optical scattering. Accordingly, using hereafter the Heisenberg picture, the general field-matter input state is of the form $|\Psi\rangle = F^{(\text{in})}(\hat{a}_s^\dagger, \hat{a}_e^\dagger)|0\rangle$, where $|0\rangle$ is the normalized

polariton vacuum state and $F^{(\text{in})}$ is a functional accounting for the ingoing s polaritons coming from the far field and the ingoing e polaritons stored in the object volume at $t = -\infty$. Since in the Heisenberg picture the state does not evolve, the output state is obtained by expressing $|\Psi\rangle$ in terms of the outgoing polariton creation operators, i.e., by substituting the Hermitian conjugates of Eq. (A11) into the above expression of $|\Psi\rangle$, thus getting $|\Psi\rangle = F^{(\text{out})}(\hat{A}_s^\dagger, \hat{A}_e^\dagger)|0\rangle$, where the obtained functional $F^{(\text{out})}$ describes the outgoing s polaritons traveling toward the far field and the outgoing e polaritons stored in the object volume at $t = +\infty$. Note that, since the scatterer object has finite size, s polaritons in the far field coincide with standard photons of vacuum quantum electrodynamics; however, we hereafter use the nomenclature of ingoing and outgoing s polaritons (instead of photons) as a reminder of the underlying dispersive nature of the scatterer object.

APPENDIX B: CLASSICAL ELECTROMAGNETIC SCATTERING BY A HOMOGENEOUS LOSSY SPHERE

In this Appendix, we evaluate the scattering coefficient $s_{\mathbf{n}\Lambda}^{\mathbf{N}\Lambda}(\omega)$ and the power scattering and absorption coefficients $S_{\mathbf{n}\Lambda}^{\mathbf{N}\Lambda}(\omega)$ and $A_{\mathbf{n}\Lambda}^{\mathbf{N}\Lambda}(\omega)$ for a homogenous lossy sphere of radius a and dielectric permittivity ϵ_ω^V ; the overall dielectric permittivity is $\epsilon_\omega(\mathbf{s}) = H(s-a) + H(a-s)\epsilon_\omega^V$, where $s = |\mathbf{s}|$ and H is the Heaviside step function. By resorting to the well-known Mie analytical treatment of plane-wave scattering by a homogeneous sphere [88], the modal dyadics $\mathcal{F}_\omega(\mathbf{s}|\mathbf{n})$ satisfying the boundary-value problem in the second row of Eq. (A1) is seen to be given by

$$\begin{aligned} \mathcal{F}_\omega(\mathbf{s}|\mathbf{n}) &= H(s-a)e^{ik_\omega\mathbf{s}\cdot\mathbf{n}}\mathcal{I}_\mathbf{n} + H(s-a)4\pi \sum_{n=1}^{\infty} \sum_{m=-n}^n i^n \\ &\times \left\{ -\frac{a_{n\omega}}{ik_\omega} \nabla_s \times \left[h_n^{(1)}(k_\omega s) \mathbf{X}_{nm} \left(\frac{\mathbf{s}}{s} \right) \right] \mathbf{n} \times \mathbf{X}_{nm}^*(\mathbf{n}) - b_{n\omega} h_n^{(1)}(k_\omega s) \mathbf{X}_{nm} \left(\frac{\mathbf{s}}{s} \right) \mathbf{X}_{nm}^*(\mathbf{n}) \right\} \\ &+ H(a-s)4\pi \sum_{n=1}^{\infty} \sum_{m=-n}^n i^n \left\{ \frac{d_{n\omega}}{ik_\omega^V} \nabla_s \times \left[j_n(k_\omega^V s) \mathbf{X}_{nm} \left(\frac{\mathbf{s}}{s} \right) \right] \mathbf{n} \times \mathbf{X}_{nm}^*(\mathbf{n}) + c_{n\omega} j_n(k_\omega^V s) \mathbf{X}_{nm} \left(\frac{\mathbf{s}}{s} \right) \mathbf{X}_{nm}^*(\mathbf{n}) \right\}, \end{aligned} \quad (\text{B1})$$

where $k_\omega^V = k_\omega(\epsilon_\omega^V)^{1/2}$ [with $\text{Im}(k_\omega^V) \geq 0$] is the wave number inside the sphere, $j_n(\rho)$ and $h_n^{(1)}(\rho)$ are the spherical Bessel and Hankel functions of the first kind,

$$\mathbf{X}_{nm}(\mathbf{m}) = \frac{1}{\sqrt{n(n+1)}} \frac{1}{i} \left(-\mathbf{u}_{\theta_{\mathbf{m}}} \frac{1}{\sin \theta_{\mathbf{m}}} \frac{\partial}{\partial \varphi_{\mathbf{m}}} + \mathbf{u}_{\varphi_{\mathbf{m}}} \frac{\partial}{\partial \theta_{\mathbf{m}}} \right) Y_{nm}(\theta_{\mathbf{m}}, \varphi_{\mathbf{m}}) \quad (\text{B2})$$

is the vector spherical harmonic associated with the scalar spherical harmonic Y_{nm} , and

$$\begin{aligned} \begin{pmatrix} a_{n\omega} \\ d_{n\omega} \end{pmatrix} &= \frac{\begin{pmatrix} j_n(k_\omega^V a) \frac{\partial}{\partial a} [a j_n(k_\omega a)] - \frac{1}{\epsilon_\omega^V} j_n(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)] \\ j_n(k_\omega^V a) \frac{\partial}{\partial a} [a h_n^{(1)}(k_\omega^V a)] - h_n^{(1)}(k_\omega^V a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)] \end{pmatrix}}{j_n(k_\omega^V a) \frac{\partial}{\partial a} [a h_n^{(1)}(k_\omega a)] - \frac{1}{\epsilon_\omega^V} h_n^{(1)}(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)]}, \\ \begin{pmatrix} b_{n\omega} \\ c_{n\omega} \end{pmatrix} &= \frac{\begin{pmatrix} j_n(k_\omega^V a) \frac{\partial}{\partial a} [a j_n(k_\omega a)] - j_n(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)] \\ j_n(k_\omega a) \frac{\partial}{\partial a} [a h_n^{(1)}(k_\omega a)] - h_n^{(1)}(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega a)] \end{pmatrix}}{j_n(k_\omega^V a) \frac{\partial}{\partial a} [a h_n^{(1)}(k_\omega a)] - h_n^{(1)}(k_\omega a) \frac{\partial}{\partial a} [a j_n(k_\omega^V a)]} \end{aligned} \quad (\text{B3})$$

are the standard Mie coefficients [81]. As a consistency check of Eq. (B1), note that in the case where there is no scattering sphere, i.e., $\varepsilon_\omega^V = 1$, the Mie coefficients become $a_{n\omega} = b_{n\omega} = 0$ and $d_{n\omega} = c_{n\omega} = 1$ so that the modal dyadics reduces to

$$\mathcal{F}_\omega(\mathbf{s}|\mathbf{n}) = e^{ik_\omega \mathbf{s} \cdot \mathbf{n}} \mathcal{I}_\mathbf{n} = 4\pi \sum_{n=1}^{\infty} \sum_{m=-n}^n i^n \left\{ \frac{1}{ik_\omega} \nabla_s \times \left[j_n(k_\omega s) \mathbf{X}_{nm} \left(\frac{\mathbf{s}}{s} \right) \right] \mathbf{n} \times \mathbf{X}_{nm}^*(\mathbf{n}) + j_n(k_\omega s) \mathbf{X}_{nm} \left(\frac{\mathbf{s}}{s} \right) \mathbf{X}_{nm}^*(\mathbf{n}) \right\}, \quad (\text{B4})$$

correctly displaying a pure dyadic plane-wave profile without scattered field (the second identity is the spherical expansion of the dyadic plane wave). It is worth pointing out that the Mie coefficients satisfy the remarkable relations

$$\begin{aligned} \frac{\text{Re}(a_{n\omega}) - |a_{n\omega}|^2}{|d_{n\omega}|^2} &= (k_\omega a) \text{Im} \left\{ \frac{k_\omega^V}{k_\omega^{V*}} j_n(k_\omega^V a) \frac{\partial}{\partial a} [a j_n^*(k_\omega^V a)] \right\}, \\ \frac{\text{Re}(b_{n\omega}) - |b_{n\omega}|^2}{|c_{n\omega}|^2} &= (k_\omega a) \text{Im} \left\{ j_n(k_\omega^V a) \frac{\partial}{\partial a} [a j_n^*(k_\omega^V a)] \right\}, \end{aligned} \quad (\text{B5})$$

whose relevance stems from the fact that their RHSs vanish when $\text{Im}(k_\omega^V) = 0$ so that we conclude that the Mie coefficients of a transparent sphere are such that $\text{Re}(a_{n\omega}) = |a_{n\omega}|^2$ and $\text{Re}(b_{n\omega}) = |b_{n\omega}|^2$.

By exploiting the asymptotic behavior of the spherical Hankel function, $h_n^{(1)}(\rho) \rho \rightarrow +\infty (-i)^{n+1} \frac{e^{i\rho}}{\rho}$, the asymptotic expansion of the modal dyadic in Eq. (B1) is straightforwardly seen to be

$$\mathcal{F}_\omega(s\mathbf{N}|\mathbf{n}) \underset{s \rightarrow +\infty}{\approx} e^{i(k_\omega s)\mathbf{N} \cdot \mathbf{n}} \mathcal{I}_\mathbf{n} + \frac{e^{ik_\omega s}}{s} \frac{4\pi i}{k_\omega} \sum_{n=1}^{\infty} \sum_{m=-n}^n [a_{n\omega} \mathbf{N} \times \mathbf{X}_{nm}(\mathbf{N}) \mathbf{n} \times \mathbf{X}_{nm}^*(\mathbf{n}) + b_{n\omega} \mathbf{X}_{nm}(\mathbf{N}) \mathbf{X}_{nm}^*(\mathbf{n})], \quad (\text{B6})$$

whose comparison with the general asymptotic relation $\mathcal{F}_\omega(s\mathbf{N}|\mathbf{n}) \underset{s \rightarrow +\infty}{\approx} e^{i(k_\omega s)\mathbf{N} \cdot \mathbf{n}} \mathcal{I}_\mathbf{n} + \frac{e^{ik_\omega s}}{s} \mathcal{S}_\omega(\mathbf{N}|\mathbf{n})$ [see the second of Eq. (A1)] directly provides the scattering dyadic

$$\mathcal{S}_\omega(\mathbf{N}|\mathbf{n}) = \frac{4\pi i}{k_\omega} \sum_{n=1}^{\infty} \sum_{m=-n}^n [a_{n\omega} \mathbf{N} \times \mathbf{X}_{nm}(\mathbf{N}) \mathbf{n} \times \mathbf{X}_{nm}^*(\mathbf{n}) + b_{n\omega} \mathbf{X}_{nm}(\mathbf{N}) \mathbf{X}_{nm}^*(\mathbf{n})], \quad (\text{B7})$$

which, for notational convenience, we write as

$$\mathcal{S}_\omega(\mathbf{N}|\mathbf{n}) = \frac{4\pi i}{k_\omega} \sum_{n=1}^{\infty} (a_{n\omega} \quad b_{n\omega}) \sum_{m=-n}^n \begin{pmatrix} \mathbf{N} \times \mathbf{X}_{nm}(\mathbf{N}) \mathbf{n} \times \mathbf{X}_{nm}^*(\mathbf{n}) \\ \mathbf{X}_{nm}(\mathbf{N}) \mathbf{X}_{nm}^*(\mathbf{n}) \end{pmatrix}. \quad (\text{B8})$$

The orthogonality properties of the vector spherical harmonics

$$\begin{aligned} \int d\mathbf{m} \mathbf{X}_{n'm'}^*(\mathbf{m}) \cdot \mathbf{X}_{nm}(\mathbf{m}) &= \delta_{nn'} \delta_{mm'}, \\ \int d\mathbf{m} \mathbf{X}_{n'm'}^*(\mathbf{m}) \cdot [\mathbf{m} \times \mathbf{X}_{nm}(\mathbf{m})] &= 0, \\ \int d\mathbf{m} [\mathbf{m} \times \mathbf{X}_{n'm'}(\mathbf{m})]^* \cdot [\mathbf{m} \times \mathbf{X}_{nm}(\mathbf{m})] &= \delta_{nn'} \delta_{mm'} \end{aligned} \quad (\text{B9})$$

enable us to easily prove that

$$\int d\mathbf{N} \mathcal{S}_\omega^{T*}(\mathbf{N}|\mathbf{n}) \cdot \mathcal{S}_\omega(\mathbf{N}|\mathbf{n}') = \left(\frac{4\pi}{k_\omega} \right)^2 \sum_{n=1}^{\infty} (|a_{n\omega}|^2 \quad |b_{n\omega}|^2) \sum_{m=-n}^n \begin{pmatrix} \mathbf{n} \times \mathbf{X}_{nm}(\mathbf{n}) \mathbf{n}' \times \mathbf{X}_{nm}^*(\mathbf{n}') \\ \mathbf{X}_{nm}(\mathbf{n}) \mathbf{X}_{nm}^*(\mathbf{n}') \end{pmatrix}. \quad (\text{B10})$$

Besides, after inserting Eqs. (B8) and (B10) into Eq. (19) of the main text, we get

$$\int d^3 \mathbf{R} \mathcal{A}_{\omega es}^{T*}(\mathbf{R}|\mathbf{n}) \cdot \mathcal{A}_{\omega es}(\mathbf{R}|\mathbf{n}') = 4 \sum_{n=1}^{\infty} (\text{Re}(a_{n\omega}) - |a_{n\omega}|^2 \quad \text{Re}(b_{n\omega}) - |b_{n\omega}|^2) \sum_{m=-n}^n \begin{pmatrix} \mathbf{n} \times \mathbf{X}_{nm}(\mathbf{n}) \mathbf{n}' \times \mathbf{X}_{nm}^*(\mathbf{n}') \\ \mathbf{X}_{nm}(\mathbf{n}) \mathbf{X}_{nm}^*(\mathbf{n}') \end{pmatrix}. \quad (\text{B11})$$

Now the addition theorem for spherical harmonics, $\sum_{m=-n}^n Y_{nm}(\theta_\mathbf{n}, \varphi_\mathbf{n}) Y_{nm}^*(\theta_{\mathbf{n}'}, \varphi_{\mathbf{n}'}) = \frac{2n+1}{4\pi} P_n(\mathbf{n} \cdot \mathbf{n}')$, where $P_n(\xi)$ is the Legendre polynomial, enables us to prove the dyadic addition formulas for the vector spherical harmonics:

$$\sum_{m=-n}^n \begin{pmatrix} \mathbf{m} \times \mathbf{X}_{nm}(\mathbf{m}) \mathbf{n} \times \mathbf{X}_{nm}^*(\mathbf{n}) \\ \mathbf{X}_{nm}(\mathbf{m}) \mathbf{X}_{nm}^*(\mathbf{n}) \end{pmatrix} = \frac{1}{4\pi} \begin{pmatrix} M_n^{11}(\mathbf{m} \cdot \mathbf{n}) & M_n^{12}(\mathbf{m} \cdot \mathbf{n}) \\ M_n^{21}(\mathbf{m} \cdot \mathbf{n}) & M_n^{22}(\mathbf{m} \cdot \mathbf{n}) \end{pmatrix} \begin{pmatrix} \mathcal{I}_\mathbf{m} \cdot \mathcal{I}_\mathbf{n} \\ \mathcal{I}_\mathbf{m} \cdot \mathbf{n} \mathbf{m} \cdot \mathcal{I}_\mathbf{n} \end{pmatrix}, \quad (\text{B12})$$

where

$$\begin{pmatrix} M_n^{11}(\xi) & M_n^{12}(\xi) \\ M_n^{21}(\xi) & M_n^{22}(\xi) \end{pmatrix} = \frac{2n+1}{n(n+1)} \begin{pmatrix} P_n'(\xi) & P_n''(\xi) \\ -(1-\xi^2)P_n''(\xi) + \xi P_n'(\xi) & -\xi P_n''(\xi) - P_n'(\xi) \end{pmatrix}, \quad (\text{B13})$$

which, when inserted into Eqs. (B8), (B10), and (B11), yields

$$\begin{aligned} \mathcal{S}_\omega(\mathbf{N}|\mathbf{n}) &= \frac{i}{k_\omega} \sum_{n=1}^{\infty} (a_{n\omega} \quad b_{n\omega}) \begin{pmatrix} M_n^{11}(\mathbf{N} \cdot \mathbf{n}) & M_n^{12}(\mathbf{N} \cdot \mathbf{n}) \\ M_n^{21}(\mathbf{N} \cdot \mathbf{n}) & M_n^{22}(\mathbf{N} \cdot \mathbf{n}) \end{pmatrix} \begin{pmatrix} \mathcal{I}_\mathbf{N} \cdot \mathcal{I}_\mathbf{n} \\ \mathcal{I}_\mathbf{N} \cdot \mathbf{nN} \cdot \mathcal{I}_\mathbf{n} \end{pmatrix}, \\ \int d\mathbf{O}_\mathbf{N} \mathcal{S}_\omega^{T*}(\mathbf{N}|\mathbf{n}) \cdot \mathcal{S}_\omega(\mathbf{N}|\mathbf{n}') &= \frac{4\pi}{k_\omega^2} \sum_{n=1}^{\infty} (|a_{n\omega}|^2 \quad |b_{n\omega}|^2) \begin{pmatrix} M_n^{11}(\mathbf{n} \cdot \mathbf{n}') & M_n^{12}(\mathbf{n} \cdot \mathbf{n}') \\ M_n^{21}(\mathbf{n} \cdot \mathbf{n}') & M_n^{22}(\mathbf{n} \cdot \mathbf{n}') \end{pmatrix} \begin{pmatrix} \mathcal{I}_\mathbf{n} \cdot \mathcal{I}_{\mathbf{n}'} \\ \mathcal{I}_\mathbf{n} \cdot \mathbf{n}'\mathbf{n} \cdot \mathcal{I}_{\mathbf{n}'} \end{pmatrix}, \\ \int d^3\mathbf{R} \mathcal{A}_{\omega\text{es}}^{T*}(\mathbf{R}|\mathbf{n}) \cdot \mathcal{A}_{\omega\text{es}}(\mathbf{R}|\mathbf{n}') &= \frac{1}{\pi} \sum_{n=1}^{\infty} (\text{Re}(a_{n\omega}) - |a_{n\omega}|^2 \quad \text{Re}(b_{n\omega}) - |b_{n\omega}|^2) \begin{pmatrix} M_n^{11}(\mathbf{n} \cdot \mathbf{n}') & M_n^{12}(\mathbf{n} \cdot \mathbf{n}') \\ M_n^{21}(\mathbf{n} \cdot \mathbf{n}') & M_n^{22}(\mathbf{n} \cdot \mathbf{n}') \end{pmatrix} \begin{pmatrix} \mathcal{I}_\mathbf{n} \cdot \mathcal{I}_{\mathbf{n}'} \\ \mathcal{I}_\mathbf{n} \cdot \mathbf{n}'\mathbf{n} \cdot \mathcal{I}_{\mathbf{n}'} \end{pmatrix}. \end{aligned} \quad (\text{B14})$$

Exploiting such dyadic expressions to evaluate the scattering coefficient $s_{\mathbf{n}\lambda}^{\text{N}\Lambda}(\omega)$ in the first of Eq. (9) and the power scattering and absorption coefficients $S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega)$ and $A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega)$ of Eq. (15), bearing in mind that $\mathcal{I}_\mathbf{m} \cdot \mathbf{e}_{\mathbf{m}\lambda} = \mathbf{e}_{\mathbf{m}\lambda}$, we get

$$\begin{aligned} s_{\mathbf{n}\lambda}^{\text{N}\Lambda}(\omega) &= \frac{1}{\pi} \sum_{n=1}^{\infty} \begin{pmatrix} -\frac{1}{2}a_{n\omega} & -\frac{1}{2}b_{n\omega} \end{pmatrix} \begin{pmatrix} M_n^{11}(\mathbf{N} \cdot \mathbf{n}) & M_n^{12}(\mathbf{N} \cdot \mathbf{n}) \\ M_n^{21}(\mathbf{N} \cdot \mathbf{n}) & M_n^{22}(\mathbf{N} \cdot \mathbf{n}) \end{pmatrix} \begin{pmatrix} \mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathbf{e}_{\mathbf{n}\lambda} \\ \mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathbf{nN} \cdot \mathbf{e}_{\mathbf{n}\lambda} \end{pmatrix}, \\ S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega) &= \frac{1}{\pi} \sum_{n=1}^{\infty} (|a_{n\omega}|^2 \quad |b_{n\omega}|^2) \begin{pmatrix} M_n^{11}(\mathbf{n} \cdot \mathbf{n}') & M_n^{12}(\mathbf{n} \cdot \mathbf{n}') \\ M_n^{21}(\mathbf{n} \cdot \mathbf{n}') & M_n^{22}(\mathbf{n} \cdot \mathbf{n}') \end{pmatrix} \begin{pmatrix} \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \\ \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{n}'\mathbf{n} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \end{pmatrix}, \\ A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega) &= \frac{1}{\pi} \sum_{n=1}^{\infty} (\text{Re}(a_{n\omega}) - |a_{n\omega}|^2 \quad \text{Re}(b_{n\omega}) - |b_{n\omega}|^2) \begin{pmatrix} M_n^{11}(\mathbf{n} \cdot \mathbf{n}') & M_n^{12}(\mathbf{n} \cdot \mathbf{n}') \\ M_n^{21}(\mathbf{n} \cdot \mathbf{n}') & M_n^{22}(\mathbf{n} \cdot \mathbf{n}') \end{pmatrix} \begin{pmatrix} \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \\ \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{n}'\mathbf{n} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \end{pmatrix}, \end{aligned} \quad (\text{B15})$$

which we rewrite in the more concise and expressive form

$$\begin{aligned} s_{\mathbf{n}\lambda}^{\text{N}\Lambda}(\omega) &= \mathbf{J}_{\mathbf{N}\cdot\mathbf{n}} \left\{ \begin{pmatrix} -\frac{1}{2}a_{n\omega} & -\frac{1}{2}b_{n\omega} \end{pmatrix} \right\} \begin{pmatrix} \mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathbf{e}_{\mathbf{n}\lambda} \\ \mathbf{e}_{\mathbf{N}\Lambda} \cdot \mathbf{nN} \cdot \mathbf{e}_{\mathbf{n}\lambda} \end{pmatrix}, \\ S_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega) &= \mathbf{J}_{\mathbf{n}\cdot\mathbf{n}'} \left\{ (|a_{n\omega}|^2 \quad |b_{n\omega}|^2) \right\} \begin{pmatrix} \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \\ \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{n}'\mathbf{n} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \end{pmatrix}, \\ A_{\mathbf{n}'\lambda'}^{\mathbf{n}\lambda}(\omega) &= \mathbf{J}_{\mathbf{n}\cdot\mathbf{n}'} \left\{ (\text{Re}(a_{n\omega}) - |a_{n\omega}|^2 \quad \text{Re}(b_{n\omega}) - |b_{n\omega}|^2) \right\} \begin{pmatrix} \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \\ \mathbf{e}_{\mathbf{n}\lambda} \cdot \mathbf{n}'\mathbf{n} \cdot \mathbf{e}_{\mathbf{n}'\lambda'} \end{pmatrix}, \end{aligned} \quad (\text{B16})$$

where

$$\mathbf{J}_\xi \{ (p_n \quad q_m) \} = \frac{1}{\pi} \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (p_n \quad q_m) \begin{pmatrix} P'_n(\xi) & P''_n(\xi) \\ -(1-\xi^2)P''_n(\xi) + \xi P'_n(\xi) & -\xi P''_n(\xi) - P'_n(\xi) \end{pmatrix}. \quad (\text{B17})$$

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