Parametric Mie resonances and Kerker effects in time-modulated scatterers

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We provide a theoretical description of light scattering by a spherical particle whose permittivity is modulated in time at twice the frequency of the incident light. Such a particle acts as a finite-sized photonic time crystal and, despite its sub-wavelength spatial extent, can host optical parametric amplification. Conditions of parametric Mie resonances in the sphere are derived. We show that control of the temporal modulation strength provides a qualitatively new route to tailor the far-field pattern of a scatterer. Two characteristic spheres are designed, providing amplification of scattered light and satisfying nearly ideally the first and second Kerker scattering conditions.

Sub-wavelength high-index dielectric resonators provide a versatile platform for light control at the nanoscale. These resonators can support strong light localization described by multipolar Mie-type resonances [1,6]. The resonant modes are generated by the volumetric distribution of displacement currents and can be of electric or magnetic kinds. A remarkable feature of Mie-type scattering lays in the possibility to spectrally overlap several multipolar modes for engineering complex scattering patterns. In particular, as it was originally suggested by M. Kerker et al., spherical particles with properly tuned electric and magnetic responses can exhibit zero light scattering either in the backward or forward direction [7]. Subsequently, these scattering regimes are referred to as the so-called first and second Kerker effects, respectively. During the last few years, various generalizations of the Kerker effects and generic scattering engineering through multimodal interference led to multiple applications in nanophotonics, including wavefront manipulations for metasurfaces [8], bound states in the continuum [9,10], nonradiating anapole modes [11,12], and directional spontaneous parametric down-conversion [13,14] among many others.

Most of the previous works on Mie-type scatterers concentrated on time-invariant particles whose permittivity does not change in time. The time variation of material properties unlocks a new dimension of control in electromagnetic systems [15,16]. Recently, a wide range of novel optical effects was suggested based on time-varying materials, such as photonic time crystals [17,22], temporal discontinuities [23-25], effective magnetic field for photons [26], optically induced negative refraction [27], synthetic dimensions [28], etc. The temporal material modulation has the potential to dramatically extend both conceptual and applied aspects of Mie-type scattering [29-30]. However, to date this area of research has remained essentially unexplored.

In this Letter, we analyse light scattering by a sphere whose permittivity is modulated at twice the frequency of the incident light, which corresponds to the case of parametric excitation. Based on Floquet Mie theory and the temporal coupled mode theory, we demonstrate that such a sphere, despite its sub-wavelength spatial extent, hosts parametric Mie resonances. It is revealed that varying the strength of the temporal modulations provides a qualitatively new route to control directional scattering by the sphere. We design two characteristic examples of parametric scatterers with directional scattering patterns possessing sharp dips in the backward and/or forward directions. A related effect of parametric amplification in spherical scatterers with the second-order nonlinearity was recently reported in Ref. [31], however, simultaneous far-field pattern engineering was not demonstrated.

We consider a sphere located at the center of the coordinate system (see Fig. 1). The material of the sphere without modulations is described by a single-pole Lorentz-Drude dispersion model with the stationary relative permittivity function given by 

\[ \varepsilon_{st}(\omega) = 1 + \omega_p^2/(\omega_i^2 - \omega^2 - i\gamma\omega), \]

where \( \gamma \) is the damping factor and \( \omega_i \) the resonance frequency. In what follows, we choose \( \omega_p = \sqrt{N_0 q_e^2/m_e \varepsilon_0} = 3.5\omega_r \), where \( q_e \) and \( m_e \) are the electron charge and mass, respectively, and \( \varepsilon_0 \) is the vacuum permittivity. Parameter \( N_0 \) is the time-averaged bulk carrier density. The temporal variation of the sphere’s permittivity \( \varepsilon \) is assumed to be via the modulation of the charge carrier density of the form \( N(t) = N_0 (1 + M \cos \omega_m t) \) (see Sec. 1 of the Supplemental Material [32]), where \( M \) is the modulation strength and \( \omega_m \) is the modulation frequency. In what follows, we choose a regime of relatively low dispersion, that is, \( \omega_m = 0.5\omega_r \). Modulation of the carrier concentration with the strength of the order of unity and \( \omega_m \)
at optical frequencies was experimentally demonstrated in several recent works [27], [33], [34].

We first find the eigenfrequencies and corresponding eigenmodes of an unbounded dispersive material with time-varying carrier concentration \( N(t) \). The wave equation of such material written for the electric field \( \mathbf{E}(\mathbf{r}, \omega) \) reads [30]

\[
\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) = k^2(\omega) \left[ \mathbf{E}(\mathbf{r}, \omega) + \int_{-\infty}^{+\infty} \chi(\omega - \omega', \omega') \mathbf{E}(\mathbf{r}, \omega') d\omega' \right].
\]

Here, \( k(\omega) = \omega/c \) is the wavenumber of free space, \( c \) is the speed of light, \( \mathbf{r} \) is the position vector, \( \chi(\omega - \omega', \omega') = \varepsilon(\omega - \omega', \omega') - \delta(\omega - \omega') \) is the generalized susceptibility that describes the polarization density at frequency \( \omega \) induced by an electric field harmonic at frequency \( \omega' \), and \( \delta(\omega - \omega') \) is the Dirac delta function. Solving the wave equation, we look for the electric field in the form \( \mathbf{E}(\mathbf{r}, \omega) = \int A(\kappa) S_\kappa(\omega) \mathbf{F}(\kappa \mathbf{r}) d\kappa \), where \( \mathbf{F}(\kappa \mathbf{r}) \) and \( S_\kappa(\omega) \) are the spatial and spectral parts of the eigenmodes, respectively, \( A(\kappa) \) is the complex modal amplitude, and \( \kappa \) is the eigen-wavenumber [30]. Due to the spherical symmetry, the spatial function can be represented as a linear combination of conventional complex-valued vector spherical harmonics (VSHs) \( \mathbf{F}^{(\ell)}_{\mu \nu}(\kappa \mathbf{r}) \), whose indices \( \mu \) and \( \nu \) stand for the angular momentum along the \( z \)-axis and the multipolar order, respectively [30], [34] Sect. 13.3. Subscript \( \alpha \) stands for one of the two labels, \( \alpha_M \) or \( \alpha_N \), and refers to magnetic or electric multipolar modes, respectively. Finally, superscript \( \ell \) takes the values “1” or “3” to refer to regular or radiating VSHs, respectively [30].

By substituting the electric field ansatz into (1), we obtain the following eigenvalue equation in the matrix form (see Sec. 1 of the Supplemental Material [32]):

\[
k^2_n (\varepsilon_{st,n} S_{\kappa,n} + \varepsilon_{dyn,n} S_{\kappa,n+1} + \varepsilon_{dyn,n} S_{\kappa,n-1}) = \kappa^2 S_{\kappa,n},
\]

where \( \varepsilon_{dyn}(\omega) = [\varepsilon_{st}(\omega) - 1] M/2 \) is the dynamic part of the relative permittivity. In (2), index \( n \) means that the corresponding function is taken at frequency \( \omega_n = \omega + n\omega_m \).

Equation (2) allows one to find a set of eigen-wavenumbers \( \kappa_q \) \((q \) is a positive integer\) for a bulk temporally modulated material at a given Floquet frequency \( \omega \) [18], as well as the matrix of weights \( S_{\kappa,n} \) of the modes with frequency \( \omega_n \) and wavenumber \( \kappa_q \). Eigenvalue equation (2) results in a band diagram with period \( \omega_m \) that corresponds to that of a photonic time crystal [18].

In Fig. 2, we plot the band diagram for the special case of the material with \( M = 0.1 \) and \( \gamma = 0 \) Hz. Since the considered material has a Lorentzian dispersion, there are two bulk plasmon-polariton bands where the real part of the permittivity is positive. These two bands are shown with blue and red lines in the figure. The first one (blue) is split by a momentum bandgap, inside which there are two modes which have purely imaginary eigenfrequencies (one attenuating and one amplifying). The amplifying mode is responsible for the parametric amplification effect in time-modulated materials and it is being excited even if the bandgap is closed by the red bands.

Next, we analyse wave phenomena in a finite-size sphere made from such a time-modulated material. First, we find the condition of optical parametric amplification. For its derivation, we will consider a separate eigenvalue problem for the electric field amplitudes across the sphere boundary with no incident field (parametric self-maintained oscillations). To find the parametric oscillation condition analytically, we consider the Floquet frequency right at the center of the momentum bandgap, that is, \( \omega = \omega_m/2 \), and exploit the weak-modulation approximation [36], which is valid in the regime of \( M \ll 1 \).

As we verified numerically, under this approximation, there are only two dominant harmonics \( \omega_0 = \omega_m/2 \) and \( \omega_1 = -\omega_m/2 \) and two dominant (lowest) momentum bands \( \kappa_1 \) and \( \kappa_2 \). In other words, the matrix of modal
weights $S_{q\nu}$ can be truncated to merely a $2 \times 2$ size with indices $q = \{1, 2\}$ and $n = \{0, -1\}$. The points with $\kappa_1$ and $\kappa_2$ are marked in the diagram of Fig. 2. Using the approximation, equation (2) can be solved analytically in a closed form (see Sec. 2 of the Supplemental Material [32]) yielding the following expressions for the momenta and modal weights for the parametric-oscillation regime:

$$\kappa_1 = \frac{\omega_m}{2c} \sqrt{\text{Re} \, \varepsilon_{st,0} - \tilde{\varepsilon}}, \quad \kappa_2 = \frac{\omega_m}{2c} \sqrt{\text{Re} \, \varepsilon_{st,0} + \tilde{\varepsilon}},$$

$$S_{q\nu} = \begin{pmatrix} \varepsilon_{\text{dyn},0}/(i \text{Im} \, \varepsilon_{st,0} - \tilde{\varepsilon}) & 1 \\ \varepsilon_{\text{dyn},0}/(i \text{Im} \, \varepsilon_{st,0} + \tilde{\varepsilon}) & 1 \end{pmatrix},$$

where "*" denotes complex conjugation and $\tilde{\varepsilon} = \sqrt{\varepsilon_{\text{dyn},0}^2 - (\text{Im} \, \varepsilon_{st,0})^2}$.

The electric field inside the sphere can be expressed using a set of VSHs as $E^\text{in}(r, \omega_n) = \sum_{\alpha, \mu, \nu} A^n_{\alpha,\mu\nu}(k_q r) S_{q\nu},$ with $A^n$ standing for amplitudes of corresponding VSHs with wavenumber $k_q$. The electric field outside the sphere (in vacuum), represented by the scattered field only, is given by $E^\text{sc}(r, \omega_n) = \sum_{\alpha, \mu, \nu} A^\text{sc}\alpha_{\alpha,\mu\nu}(\omega_n) F^{(1)}_{\alpha,\mu\nu}(k_n r).$ Substituting these expressions into the boundary conditions at the surface of the sphere with radius $R$ ($r = R \hat{r}$) and using the orthogonality relations for VSHs, we obtain the following system of equations (see Sec. 2 of the Supplemental Material [32]):

$$\sum_{q=1}^2 A^n_{\alpha,\mu\nu} S_{q\nu} \zeta^{(1)}_{\alpha,\mu}(k_q R) = A^\text{sc}\alpha_{\alpha,\mu\nu}(\omega_n) \zeta^{(3)}_{\alpha,\mu}(k_n R),$$

$$\sum_{q=1}^2 A^n_{\alpha,\mu\nu} S_{q\nu} \kappa_q \gamma^{(1)}_{\alpha,\mu}(k_q R) = A^\text{sc}\alpha_{\alpha,\mu\nu}(\omega_n) \gamma^{(3)}_{\alpha,\mu}(k_n R).$$

Here, $\zeta^{(i)}_{\alpha,\mu}$ denotes the spherical Bessel ($\iota = 1$) and Hankel ($\iota = 3$) functions of the first kind of order $\nu$. Index $\beta$ is always different from $\alpha$, that is, if $\alpha = \alpha_M$ then $\beta = \alpha_N$, and vice versa. Equations (4) must hold for each set of parameters $\{\alpha, \mu, \nu, n\}$. Writing these two equations for the two frequency harmonics $n = 0$ and $n = -1$, we finally formulate the electric field equation for the electric field amplitudes across the sphere boundary, i.e., with respect to field amplitudes $A^n_{\alpha,\mu\nu1}$, $A^n_{\alpha,\mu\nu2}$, $A^\text{sc}\alpha_{\alpha,\mu\nu}(\omega_1)$, and $A^\text{sc}\alpha_{\alpha,\mu\nu}(\omega_0)$. For the regime of parametric oscillations in the sphere (in the absence of incident waves), we are looking for the solutions with nonzero amplitudes $A^n$ and $A^\text{sc}$. Therefore, we equate the determinant of the $4 \times 4$ matrix in the eigenvalue problem to zero and solve the resulting equation with respect to the radius $R$ and the modulation strength $M$ of the sphere. Figure 3 depicts the solutions of the zero matrix determinant for electric-type ($\alpha = \alpha_N$) and magnetic-type ($\alpha = \alpha_M$) modes in the sphere with multipolar orders from $\nu = 1$ to $\nu = 5$, indicating the threshold values of the modulation strength to provide parametric oscillations. The data are plotted for $\gamma = 0$ Hz. Non-zero dissipation would lead to merely a minor change in Fig. 3 shifting all the curves to the right side. As it follows from (4), the solutions are independent of parameter $\mu$. One can observe from the plot that higher-order multipolar modes (with larger values of $R \omega_n/2c$ and higher quality factors) can host parametric oscillations at lower values of $M$. Note that, although approaching, the lines never cross the vertical axis. For example, the magnetic multipole of the order $\nu = 5$ ($\alpha = \alpha_{M5}$, green solid line) exhibits parametric oscillation at the value of $M$ as low as $2.27 \times 10^{-4}$. As one example, we also depict in the plot with the black dashed horizontal line the normalized radius which provides conventional Mie resonance for the magnetic octupole mode ($\alpha = \alpha_{M3}$) in a non-modulated sphere. As one can see, the black line lays right at the radius where the least $M$ necessary to induce a parametric resonance occurs for the corresponding multipolar mode calculated for the time-varying sphere (cusp of solid orange line).

To analyse the physics of parametric Mie resonances, we employ a temporal coupled-mode theory [37, 38]. Let us consider two coupled quasi-normal modes inside the sphere at frequencies $\pm \omega_m/2$ with the total electric field of the form $E(r, \omega) = a_1(t) e^{-i \omega_{tot} t/2} [E^\text{Mie}(r)]^* + c.c.$ Here, $a_1(t)$ and $a_2(t)$ are the slowly varying temporal envelopes of the original and time-reversed modes and $E^\text{Mie}(r)$ is the spatial mode profile. We assume that $\omega_m/2$ is close to the frequency $\omega_{\text{tot}}$ that corresponds to one of the stationary Mie resonances, that is, $\omega_{\text{tot}} = \omega_m/2 - \Delta \omega - i \gamma_{\text{tot}}$ (where $|\Delta \omega + i \gamma_{\text{tot}}| \ll \omega_m/2$). Here, $\gamma_{\text{tot}}$ is the total decay rate which includes radiation and possible dissipation losses. Starting from the wave equation in the time-modulated material, one can arrive to the following system of coupled-mode equations describing evolution of
mode envelopes $a_1(t)$ and $a_2^*(t)$ inside the sphere (see Sec. 3 of the Supplemental Material [32]):

\[
\begin{align*}
\frac{d}{dt} a_1(t) &= [i\Delta \omega - \gamma_{tot}] a_1(t) + i\eta a_2^*(t), \\
\frac{d}{dt} a_2^*(t) &= [-i\Delta \omega - \gamma_{tot}] a_2^*(t) - i\eta^* a_1(t),
\end{align*}
\]

where $\eta$ is a coupling parameter linearly proportional to modulation strength $M$. Solving system (5), we obtain the threshold value of modulation strength $M_{thr} \propto \gamma_{tot} + \frac{1}{2\gamma_{tot}} \Delta \omega^2$ for parametric amplification in the sphere. This value provides a qualitative description of the spectral lineshapes of the parametric Mie resonances in Fig. 3. Indeed, the lines represent parabolas shifted from the vertical axis by a distance proportional to the decay rate $\gamma_{tot}$. For modes with higher multipolar orders $\nu$, the decay rate due to radiation loss is smaller, which results in their closer proximity to the vertical axis in Fig. 3. By selecting points where lines intersect, we can find an optimal configuration of $M$ and $R$ which ensures simultaneous parametric amplification of two different modes. The orientation of these modes is locked when the sphere is illuminated by incident light. Thus, variation of the modulation strength $M$ provides a qualitatively new route to achieve directional scattering by the sphere.

In what follows, we consider two representative examples of parametric spheres. In both examples the sphere is illuminated by monochromatic plane waves at a frequency $\omega_{inc}$ (see Fig. 1). The incident frequency is slightly shifted away from $\omega_m/2$ so that we can achieve finite and controllable amplification and use the harmonic-field analysis. From a practical point of view, the amplification can be locked-in to frequency $\omega_{inc}$ instead of $\omega_m/2$ if temporal modulations occur while the sphere is illuminated by the incident light [39].

For the first example, we consider a sphere configuration with $M = 0.68$ and $R = 1.048 \frac{\omega_m}{\omega_{inc}}$, marked by point A in Fig. 3. The configuration corresponds to the first parametric resonance crossing of the electric and magnetic dipole modes. Since contours in Fig. 3 were plotted under the approximation of $M \ll 1$, for finding the exact coordinates of point A, we calculated the contours considering a large number of frequency harmonics (see Sec. 4 in Supplemental Material [32]). In the present and the following examples, we chose $\gamma = 0$. We excite the sphere by incident light at $\omega_{inc} = 0.498 \omega_m$. Figure 3(a) depicts the scattered far-field pattern at frequency $\omega_{inc}$. The pattern is unidirectional, revealing zero backward scattering due to close fulfillment of the first Kerker condition [7]. The condition implies that the electric and magnetic modes in the sphere have approximately same amplitudes and phases. We were able to reach such a balance by fine adjustments of parameters $M$, $R$, and $\omega_{inc}$. The scattering and absorption cross sections in this example are $C_{sca}/C_{geom} = 2629.2$ and $C_{abs}/C_{geom} = -2627.5$, where $C_{geom} = \pi R^2$ and negative sign of $C_{abs}$ implies the activity of the modulated sphere. Clearly, the scattering cross section largely exceeds that of the same sphere without temporal modulations (for which case $C_{sca}^{stat}/C_{geom} = 5.5$ and $C_{abs}^{stat} = 0$ due to the presence of modulation (gain).

The second example is a sphere with a configuration of $M = 0.093$ and $R = 1.481 \frac{\omega_m}{\omega_{inc}}$ which coincides with the parametric resonance crossing of the electric quadrupole and magnetic octupole modes. This configuration is marked by point B in Fig. 3. Incident light at frequency $\omega_{inc} = 0.4995 \omega_m$ is scattered by the sphere with the pattern shown in Fig. 3(b). Such a pattern has a sharp dip in both the backward and the forward direction. Note that, whereas the electric and magnetic dipoles have opposite parity symmetry, ensuring the first Kerker condition, the electric quadrupole and magnetic octupole have the same parity symmetry, allowing for the engineering of the second Kerker condition. The scattering and absorption cross sections are $C_{sca}/C_{geom} = 858.3$ and $C_{abs}/C_{geom} = -857.5$ (in comparison, $C_{sca}^{stat}/C_{geom} = 2.53$ and $C_{abs}^{stat} = 0$ for the stationary sphere). Surprisingly, for both considered time-modulated spheres, the conventional optical theorem [40], written for the forward scattering and extinction cross section at the fundamental frequency $\omega_{inc}$, is precisely satisfied, despite the obvious optical gain in the sphere indicated by the giant scattering cross section. The peculiar pattern in Fig. 3(b) with scattering dips in both forward and backward directions stems from the precise engineering of amplitude and phases of the two multipolar modes (see Sec. 5 of the Supplemental Material [32]).

In summary, we have explored optical parametric amplification by spherical scatterers with time-modulated permittivity. The presented two geometries highlight the fascinating opportunities of simultaneous light amplification and scattering pattern control. Such particles can find applications for designing nanoscale parametric amplifiers. Due to the directional nature of their scattering and possibility of finite amplification, one can create...
exotic non-attenuating waveguide modes and topological edge modes in a non-uniform lattice of such spheres. Moreover, the second sphere example can provide unlimited scattered power for very weak incident light signals, paving the way to novel nanoscale sensors. Such scattering will cause only a small shadow for incident light in the forward direction. Due to the symmetry of the sphere, it is possible to determine the propagation direction of the light under detection by looking at the scattering pattern. Our results can be extended to particles with other geometries and represent the first step towards parametric metasurfaces based on time-modulated scatterers.

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