The role of Purcell effect for third harmonic generation

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Abstract. Third-harmonic generation from a nonlinear layer in a dielectric resonator is considered. It is shown theoretically that the Purcell effect can significantly affect the efficiency of harmonic generation. To describe THG by complex cavities, a homogenization approach is developed. It is shown that this approach works well for modelling the emission from a nanostructured surface consisting of subwavelength-size nonlinear nanoparticles.

1. Introduction
Nonlinear optics has been attracting the interest of researchers for several decades. Numerous nonlinear optical phenomena have been discovered, the most important of them are frequency mixing, higher-harmonic generation, supercontinuum generation, optical solitons, etc., see [1–4]. The theory of new frequencies generation at the interface between linear and nonlinear media has been developed since the pioneering works by Bloembergen et al. [5]. In particular, the expressions for reflected and transmitted second-harmonic light from a thin nonlinear dielectric slab were obtained. It was demonstrated that the refraction on the boundary between media with different dielectric susceptibility will affect the intensity of the emitting radiation at higher harmonics. The possibility of enhancement of the nonlinear effects by using optical resonators, in other words by exploiting the Purcell effect, was considered in [6–10]. It is well known that the Purcell effect contributes to the rates of spontaneous emission. In the case of higher harmonics generation by nonlinear processes, the efficiency also depends on the local density of photonic states at the location of the nonlinear polarization source [11]. It is shown [12] that the Purcell effect can increase the photoluminescence in Si ring resonators by at least 50 times. There are several papers devoted to the consideration of new harmonic generation from a planar nonlinear dielectric film placed inside a dielectric resonator [13, 14]. The research on the new frequencies generation by the arrays of nonlinear nanoparticles has been recently reported in a number of papers [15–19]. The optical properties of nanoscale structures differ significantly from the properties of bulk materials because of the strong confinement of the electromagnetic field inside nanoparticles resulting in geometrical resonances. In the recent work [20], the emission of the third harmonic from dielectric (silicon) nanodisks placed on a glass layer was reported. A pronounced reshaping of the third-harmonic spectra and the increase of THG intensity by two orders of magnitude with respect to bulk silicon was explained in the terms of the interplay between the electric and magnetic dipole resonances.

2. THG from a thin nonlinear film
The main goal of the paper is to consider how the interplay of two effects, the enhancement of the pump field and the Purcell effect, influences the process of third harmonic generation. For this purpose we consider a very simple physical system consisting of a dielectric Fabry-Perot resonator with a thin
nonlinear layer. The resonator is pumped by external radiation at the fundamental frequency $\omega_1 = \Omega$. In our model a nonlinear layer has a cubic (Kerr) nonlinearity and thus the incident radiation excites the current at the frequency triple of that of the pumping wave ($\omega_3 = 3\Omega$). This current causes the emission of the third harmonic. We assume that the nonlinearity is weak and we can disregard the influence of the third harmonic on the propagation of the fundamental mode. Because of the same reason, we disregard generation of higher harmonics.

In this case the problem can be split into two parts. The first part is finding a solution for the distribution of the field at the fundamental frequency. For this we need to solve the Helmholtz equation with the appropriate boundary conditions at the infinity (no radiation coming from minus infinity and a fixed incident wave coming from plus infinity).

$$\hat{H}(\omega_1) E_1 = 0. \quad (1)$$

The second part of the problem is the solution of the Helmholtz equation for the third harmonic $E_3$ with no incident waves.

$$\hat{H}(\omega_3) E_3 = \frac{4\pi}{c^2} \omega_3^2 P_3, \quad (2)$$

where the nonlinear polarization vector is determined as $P_3 = \chi^{(3)} \left| E_1 \right|^2 E_1$. It is important to note that equation (2) is the usual linear wave equation with driving force associated with the oscillations of the polarization at the third harmonic of the fundamental frequency $\omega_3 = 3\Omega$ in the right-hand side of the equation. Let us note that the operator $\hat{H}$ depends on the frequency if dielectric permittivity $\varepsilon$ is a function of frequency.

The amplitude of the third harmonic field $E_3$ is the solution of equation (2) and can be expressed through Green functions:

$$E_3 = \int \hat{G}(r, r') f(r') dr', \quad (3)$$

where $f = \frac{4\pi}{c^2} \omega_3^2 \chi^{(3)} \left| E_3 \right|^2 E_3$. The tensor Green function for the equation (3) $\hat{G}(r, r')$ is defined as a solution of the equation:

$$\hat{H}(\omega_3) \hat{G}(r, r') = \delta(r - r') \hat{I}, \quad (4)$$

where $\hat{I}$ is the unity tensor. The important point is that the radiation at the third harmonic depends on both the distribution of the fundamental field (it gives the emitting source) and the spatial structure of the Green function (which defines the coupling of the source and the emitting wave). The latter we refer to as the Purcell effect.

To demonstrate how the Purcell effect can influence third harmonic generation, let us consider a one-dimensional model that can be easily studied analytically. The system is a Fabry-Perot resonator formed by air, a glass layer of thickness $d$ and a silicon substrate. We assume that the resonator is pumped by the radiation at normal incidence (along the $z$-axis) and its polarization is perpendicular to the plane of incidence. The schematic view of the resonator under consideration is shown in figure 1. In this system at the distance $h$ from the air-glass interface we place a very narrow layer ($\delta$-layer) of a nonlinear material with cubic nonlinearity.
According to our plan, we start with propagation of the wave at the fundamental frequency. Using the well-known transfer matrix method it is easy to obtain a solution of the equation (1) for the pumping field amplitude along the propagation axis $z$:

$$E_1(z) = \begin{cases} 
\frac{1}{q} E_0 (r_{12} e^{ik_2d} + r_{23} e^{-ik_2d}) e^{-ik_1z} \equiv R_1(z) & \text{if } z \leq 0; \\
\frac{1}{q} E_0 t_{12} e^{ik_2(d-z)} + r_{23} e^{-ik_2(d-z)} & \text{if } 0 < z \leq d; \\
\frac{1}{q} E_0 t_{12} t_{23} e^{-ik_3z} \equiv T_1(z) & \text{if } z > d,
\end{cases}$$

where $k_i$ is the wavevector in the medium with index $i$, $r_{ij} = (n_i - n_j)/(n_i + n_j)$, $t_{ij} = 2n_i/(n_i + n_j)$, index "1" corresponds to air, "2" - to glass, "3" - to silicon; and we used the notation $q = (e^{ik_2d} + r_{12} r_{23} e^{-ik_2d})$.

The reflection and transmission coefficients for the pumping wave for a Fabry-Perot resonator with glass thickness $d = 2 \mu m$ are shown in figure 2. Here, $R_1$ and $T_1$ are the reflected and transmitted parts of pump energy; the curves are plotted as a function of the fundamental frequency. It is obvious that the reflection and transmission coefficients oscillate depending on the fundamental wavelength.

It is straightforward to calculate the Green function for the third harmonic excitation. In our model, it is suggested that the nonlinear source is the $\delta$-layer located in the vicinity of the air/glass boundary at a distance $h$. Thus, it is easy to find an analytical expression for the Green function:

$$E_1(z) = \frac{1}{ik_2} \begin{cases} 
(1 - r_{12}^2)/t_{12} u / w e^{-ik_1z} & \text{if } z \leq 0; \\
u / w (e^{ik_2z} - r_{12} e^{-ik_2z}) & \text{if } 0 < z \leq z'; \\
v / w (e^{ik_2(d-z)} - r_{12} e^{-ik_2(d-z)}) & \text{if } z' < z \leq d; \\
l_{23} v / w e^{-ik_3z} & \text{if } z > d,
\end{cases}$$

Figure 1. Geometry of our problem: THG by a nonlinear layer in the Fabry-Perot.
where we used the notations: 

\[ u = (e^{ik_2(d-z')} + r_{23}e^{-ik_2(d-z')}), \]

\[ v = (e^{ik_2z'} - r_{12}e^{-ik_2z'}), \]

\[ w = (r_{23}e^{-ik_2(d-z')} - e^{ik_2(z'-d)}) (e^{ik_2z'} - r_{12}e^{-ik_2z'}) - (e^{ik_2z'} + r_{12}e^{-ik_2z'})(r_{23}e^{-ik_2(d-z')} + e^{ik_2(d-z')}). \]

An important fact is that the Green function oscillates, the oscillation period is approximately three times shorter compared to the period of the fundamental harmonic. It is also worth mentioning that, because of the dispersion, the positions of the maxima of the imaginary part of the Green function and the nonlinear polarization do not coincide. The dependence of the fundamental wave intensity at the location of the \( \delta \)-layer and the Green function \( \hat{G}(z, z'=h) \) on the propagation coordinate \( z \) are shown in the inset in figure 2. The calculations are done for the parameters \( d = 3 \) \( \mu \)m, \( \Omega = 1.25 \) \( \mu \)m.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2}
\caption{Reflection (black line) and transmission (red line) coefficients as functions of wavelength for the fundamental harmonic. The inset: Dependence of the amplitude of the fundamental wave (blue) and the Green function (green) \( \hat{G}(z, z'=h) \) for the fixed on the propagation coordinate; the fundamental frequency \( \Omega = 1 \) \( \mu \)m.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3}
\caption{The dependence of the third harmonic radiation intensity on wavelength (green curve) for the case when the nonlinear layer is situated at the air-glass interface. The blue line shows the intensity of the pump wave at the position of the layer \( E_i(z=h) \). \( d = 3 \) \( \mu \)m, \( h = 0.01 \) \( \mu \)m.}
\end{figure}

In the considered case the expression in the right-hand side of the inhomogeneous wave equations for the third harmonic can be written as

\[ f(z) = 4\pi / c^2 \omega_0^2 k^{(3)}(E_1)^3 \delta(z-h). \]

Therefore, it is easy to obtain the amplitude of the third harmonic:

\[ E_3(z) = \hat{G}(z, h) \frac{4\pi}{c^2} \omega_0^2 k^{(3)}(E_1)^3. \]  \( (7) \)

As seen from equation (7), the radiation intensity for the third harmonics is given by the product of the nonlinear polarization and the Green function. In other words, the radiation intensity is proportional to the intensity of nonlinear current and this component has its maximum when the fundamental harmonic is in resonance. In the same time, the radiation intensity is also proportional to the coupling of the current to the radiating mode. This dependence is given by the Green function. This phenomenon known as the Purcell effect makes the radiation intensity frequency-dependent with pronounced maxima at certain frequencies.
The bottom panel in figure 3 shows the dependence of the pump field intensity at the layer point \( z = h \) on wavelength. The induced nonlinear current at the third harmonic is proportional to the cube of this value. On the top panel in figure 3, the normalized radiation intensity of the third harmonics (for fixed \( z < 0 \)) is shown as a function of wavelength. It is seen that the maxima of the radiation intensity are close to the maximum of the nonlinear current. However, the radiation intensity has three maxima. In a way it can be interpreted as an analogy of Fano resonance when a wider resonance (Fabry-Perot resonance for the fundamental frequency) is modulated by sharper resonances (the Green function for the third harmonic). The additional peaks in the curve are not symmetrical due to the dispersion.

3. THG from nanostructured interface

The third-harmonic generation by nanostructured nonlinear surfaces can be described using the model discussed above. By applying the homogenization scheme, we have demonstrated the Purcell effect in THG by the discrete array of nonlinear nanodisks. We examined the silicon discs with thickness \( d_1 \) and diameter \( d_2 \) arranged at a distance \( d_1 \) from each other. Nanodisks are placed on a glass plate with a thickness \( d_4 \), laying on a silicon substrate. In our calculations, we considered thin disks \( (d_1 \ll \lambda_3/n_3) \) located at a close distance (this condition means that the period of arrangement \( d_1 + d_2 \) does not exceed the third harmonic wavelength \( \lambda_3 \)).

![Figure 4](image-url)

**Figure 4.** THG by the array of nonlinear nanodisks obtained using the homogenization procedure. Parameters of the samples are listed in the inset: the distance between discs \( d_1 = 200 \) nm; the diameter \( d_2 = 100 \) nm; the thickness \( d_3 = 20 \) nm; the thickness of the glass layer \( d_4 = 2 \) μm. The normalized third harmonic radiation intensity by the effective homogeneous layer (solid blue curve) and by the thin silicon film with thickness \( b = 1 \) nm (dashed green curve) vs wavelength. In the right panels the effective parameters of the homogeneous layer are shown.
We modeled third-harmonic generation by nanodiscs using a program CST Microwave Studio. Comparing the results of numerical calculations with analytical results for multilayered structures we can obtain the effective parameters of a homogenized layer, an example is shown in the right panels of figure 4. Then, the dependence of THG by the effective nonlinear layer can be calculated straightforwardly. It is clearly seen that the dependence of the normalized harmonic intensity for the nonlinear array of nanodisks (solid blue curve) and for homogeneous silicon layer (dashed green curve) are in good agreement. This means that theory developed in this paper is applicable to the description of the harmonic generation by nanoparticle arrays of subwavelength size.

4. Conclusion
Now we briefly summarize the main results of the paper. It has been demonstrated that the Purcell effect is of great importance for third-harmonic generation. The Fabry-Perot resonance for the fundamental frequency increases the nonlinear current when exciting the third harmonic. However, the intensity of third harmonic generation depends on the product of the current and the coupling of the current to the emitting mode. In the general case, the maximum of the driving current and the maximum of the coupling are achieved at different wavelengths. This can lead to the appearance of additional maxima on the dependence of the third harmonic power on wavelength. This can be an alternative explanation of the multi-hump intensity-wavelength dependencies observed in experiments [20] and usually attributed to the closely situated electric and magnetic dipole resonances.

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