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NONLINEAR LIGHT GENERATION IN MULTILAYERS: CAVITY EFFECTS

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Using a recently derived form of the Green function for a multilayer, a compact and transparent framework for consideration of nonlinear processes in layered systems is developed within the approximation of undepleted fundamental waves. The theory is particularly suitable for the analysis of cavity (multiple–interference) effects on nonlinear light generation in multilayers and planar cavities. This is illustrated by considering harmonic generation from a nonlinear slab embedded in a multilayer and briefly discussing cavity effects in such systems.

1. *Introduction*

The interference of multiply reflected light in a (nearly) transparent layer may cause large variations of an optical–signal intensity with layer thickness, light frequency and its propagation angle. This is well known in Raman spectroscopy of reflecting substrates overlaid by dielectric layers [1]. Recently, the same effects have also been observed in second-harmonic generation (SHG) from similar systems [2–6] and, clearly, should be observable in other nonlinear processes. From a more fundamental point of view, these multiple–interference phenomena can be considered as a manifestation of the weak "quantization" of the electromagnetic field confined in a low-finesse cavity formed by the substrate and the overlayer-air interface as the second mirror. Thus, whenever a light wave involved in a process happens to coincide with a resonant mode of such a cavity, an enhanced signal intensity (cross section) is obtained. This, quantum-optical point of view is very useful when discussing multiple-interference effects in more complex systems. In quantum optics, the use of a resonant cavity for enhancing the nonlinearly generated signal is an old idea. For example, in the search for efficient monolithic harmonic generators, considerable attention has recently been paid to cavity-embedded semiconductor multilayered systems [7–10].

To describe nonlinear light generation in a multilayer properly, one must account for all reflections of all waves involved in the process. Within the approximation that fundamental waves are essentially unaffected by the nonlinear interaction, this is usually done by calculating the relevant fields using the transfer–matrix method [11] or, mostly for lower layered systems, using the Green-function formalism [12]. Very recently a combination of these two approaches has been proposed in which the Green-function technique is used to obtain generated field in a nonlinear layer from given fundamental fields, whereas wave reflections are accounted for by means of the transfer-matrix method [13]. In this work we reconsider the theory of nonlinear processes in lagered systems and develop a compact and transparent formalism for calculating generated fields. It is based fully on the Green-function method and a recently derived compact form of the Green function for an arbitrary multilayer [14]. Since the Maxwell boundary conditions are already built in the Green function, the total field generated in a nonlinear process is obtained in this framework by an (elementary) integration. In this way, one avoids a rather cumbersome matching of the bound field and the free field necessary in the transfer–matrix method [11] or matching of the self–field and the additional field necessary in the combined method [13]. Furthermore, the Green function employed is expressed as a dyadic formed by the electric-field functions of the waves incident on the system that already account for multiple wave reflections by means of the generalized Fresnel coefficients. Since these same field functions enter into the expression for nonlinear polarization, this approach leads to a compact and transparent analytical result for the field generated in an mth-order wave-mixing process in a multilayered system involving the relevant Fresnel coefficients as input parameters. The problem of calculating the generated field therefore reduces effectively to the problem of calculating the transmission and reflection coefficients of the corresponding stacks of layers, that is, to a standard problem in optics of multilayers. This makes the theory particularly convenient for the analysis of cavity (multiple-interference) effects on nonlinear light generation in multilayers and planar cavities. As an illustration, we consider harmonic generation from a nonlinear slab embedded in a multilayer and briefly discuss cavity effects in such a system. This system resembles the configurations of the novel solid-state nonlinear cavities for harmonic generators [7–10] and is therefore of obvious interest in spectroscopy of multilayers as well as in quantum optics.

2. Theory

2.1. Fundamental fields

Consider a multilayered system whose linear properties can be described by the dielectric function $\varepsilon(\mathbf{r}, \omega)$ defined in a stepwise fashion, as depicted in Fig.

1. Denoting the (conserved) wave vector parallel to the system surfaces by $\mathbf{k} = (k_x, k_y)$, the wave vector of an upward (downward) propagating wave in an lth layer is written as $\mathbf{K}_l^{\pm} = \mathbf{k} \pm \beta_l \hat{\mathbf{z}}$, where

$$\beta_l = \sqrt{k_l^2 - k^2} = \beta_l' + i\beta_l'', \quad \beta_l' \geq 0,\ \beta_l'' \geq 0,$$

$$k_l(\omega) \equiv \sqrt{\varepsilon_l(\omega)}\tilde{k} = [\eta_l(\omega) + i\kappa_l(\omega)]\tilde{k} = n_l(\omega)\tilde{k}, \tag{1}$$

with η_l and κ_l being the refractive index and the extinction coefficient of the layer, respectively, and $\tilde{k} = \omega/c$. For a transparent layer (k_l is real), propagating waves ($k \leq k_l$) may also be described by the corresponding propagation angles ϑ_l, so that $\beta_l = k_l \cos\vartheta_l$. With this notation, a $q = p$- or a $q = s$-polarized linearly propagating plane wave incident upon the system from its upper (n) or lower (0) side (assumed transparent) can be written in the form (the factor $\exp(-i\omega t)$ and the added complex-conjugate field are understood)

$$\mathbf{E}_{\mathbf{k}q}^{\nu\ \mathrm{inc}}(\mathbf{r}, \omega) = \boldsymbol{\mathcal{E}}_q^{\nu}(\mathbf{k}, \omega; z) E_q e^{\mathrm{i}\mathbf{k}\cdot\boldsymbol{\rho}}, \quad \nu = n,\ 0, \tag{2}$$

where $\boldsymbol{\rho} = (x, y)$ and E_q is related to the intensity of the beam. In a representation in which $0 < z < d_l$ in an $l{\neq}0, n$ layer, whereas $-\infty < z < 0$ in the bottom (0) and $0 < z < \infty$ in the top (n) layer, the functions $\boldsymbol{\mathcal{E}}_q^{\nu}$ are given [14]

$$\boldsymbol{\mathcal{E}}_{ql}^{n(0)}(\mathbf{k}, \omega; z) = \frac{t_{\nu/l}^q e^{\mathrm{i}\beta_l d_l}}{D_{ql}} \boldsymbol{\mathcal{E}}_{ql}^{\overset{<}{>}}(\mathbf{k}, \omega; z), \quad D_{ql} = 1 - r_{l-}^q r_{l+}^q e^{2i\beta_l d_l},$$

$$\boldsymbol{\mathcal{E}}_{ql}^{\overset{<}{>}}(\mathbf{k}, \omega; z) = \hat{\mathbf{e}}_{ql}^{\mp} e^{-\mathrm{i}\beta_l z_{\mp}} + r_{l\mp}^q \hat{\mathbf{e}}_{ql}^{\pm} e^{\mathrm{i}\beta_l z_{\mp}}, \quad z_- \equiv z,\ z_+ \equiv d_l - z,$$

$$\hat{\mathbf{e}}_{pl}^{\pm}(\mathbf{k}) = \frac{1}{k_l}(\mp\beta_l \hat{\mathbf{k}} + k\hat{\mathbf{z}}), \quad \hat{\mathbf{e}}_{sl}^{\pm}(\mathbf{k}) = \hat{\mathbf{k}} \times \hat{\mathbf{z}}, \tag{3}$$

for $l = 0, \dots n$. Here the upper (lower) sign corresponds to $\boldsymbol{\mathcal{E}}_{ql}^{n}$ ($\boldsymbol{\mathcal{E}}_{ql}^{0}$), $\hat{\mathbf{e}}_{ql}^{\pm}$ are the unit (complex) polarization vectors associated with the upward (downward) propagating wave in the lth layer, whereas $t_{n(0)/l}^q$ and $r_{l\pm}^q \equiv r_{l/n(0)}^q$ are, respectively, the transmission and reflection coefficients of the upper (lower) stack of layers bounding the layer l. For the outmost layers, $l = n$ (0), one must let $t_{n/n}^q$ ($t_{0/0}^q$) $= 1$ and d_n (d_0) $= 0$ in Eq. (3) since these quantities appear only formally. Also, obviously, $r_{n+}^q = 0$ and $r_{0-}^q = 0$. The remaining Fresnel coefficients satisfy

$$t_{i/j/k}^q = \frac{1}{D_{qj}} t_{i/j}^q t_{j/k}^q e^{\mathrm{i}\beta_j d_j} = \frac{\beta_i}{\beta_k} t_{k/j/i}^q, \tag{4a}$$

$$\tag{4}$$

$$r_{i/j/k}^q = \frac{1}{D_{qj}} \left[r_{i/j}^q + (t_{i/j}^q t_{j/i}^q - r_{i/j}^q r_{j/i}^q) r_{j/k}^q e^{2i\beta_j d_j} \right], \tag{4b}$$

and, for a single $i - j$ interface, reduce to $t^q_{ij} = \sqrt{\gamma^q_{ij}}(1 + r^q_{ij})$ and $r^q_{ij} = (\beta_i - \gamma^q_{ij}\beta_j)/(\beta_i + \gamma^q_{ij}\beta_j)$, where $\gamma^p_{ij} = \varepsilon_i/\varepsilon_j$ and $\gamma^s_{ij} = 1$, respectively. They can be calculated by using the above recurrences or by using any other suitable algorithm, e.g., the transfer matrix–method.

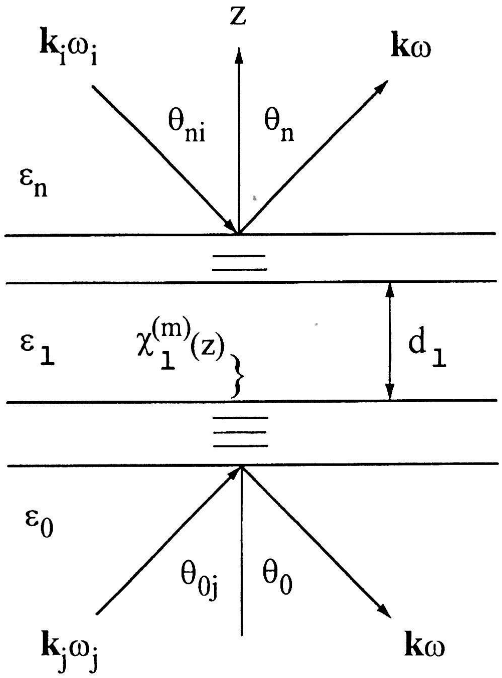

Fig. 1. System considered schematically. $\varepsilon_l = n^2_l$ are the (complex) dielectric functions of the layers. Except the bottom or the top layer, all other layers may, in principle, be nonlinear with the susceptibilities $\chi^{(m)}_l(z)$.

2.2. Nonlinear polarization

Assuming that fundamental waves propagate essentially linearly and using Eq. (2), one can write the polarization leading to an mth–order nonlinear process as [15]

$$\mathbf{P}^{NL}(\mathbf{r},\omega) = \mathbf{P}^{(m)}(\mathbf{k},\omega;z)e^{i\mathbf{k}\cdot\boldsymbol{\rho}}, \quad \omega = \sum_{i=1}^{m}(\pm\omega_i),\ \mathbf{k} = \sum_{i=1}^{m}(\pm\mathbf{k}_i),$$

$$\mathbf{P}^{(m)}(\mathbf{k},\omega;z) = \chi^{(m)}(z) : [\boldsymbol{\mathcal{E}}^{\nu_i}_{q_i}(\mathbf{k}_i,\omega_i;z)E_{q_i}]^m, \tag{5}$$

where $\chi^{(m)}$ is the corresponding susceptibility tensor. Here we have used the notation $[\boldsymbol{\mathcal{E}}_i]^m \equiv \boldsymbol{\mathcal{E}}_1(\boldsymbol{\mathcal{E}}_1^*)\ldots\boldsymbol{\mathcal{E}}_m(\boldsymbol{\mathcal{E}}_m^*)$, where $\boldsymbol{\mathcal{E}}_i^*$ appears with the $-\omega_i$ term. Similarly, for scalar quantities we write $[a_i]^m \equiv a_1(a_1^*)\ldots a_m(a_m^*)$.

2.3. Generated field

Using Eq. (5), the generated field in the system is given by [12]

$$\mathbf{E}(\mathbf{r},\omega) = e^{\mathrm{i}\mathbf{k}\cdot\boldsymbol{\rho}}\,(\frac{\omega}{c})^2\int \mathrm{d}z'\overleftrightarrow{\mathbf{G}}(\mathbf{k},\omega;z,z')\cdot\mathbf{P}^{(m)}(\mathbf{k},\omega;z'), \tag{6}$$

with $\overleftrightarrow{\mathbf{G}}(\mathbf{k},\omega;z,z')$ being the appropriate Fourier transform of the Green function for the system. $\overleftrightarrow{\mathbf{G}}(\mathbf{k},\omega;z,z')$ is represented by the elements $\overleftrightarrow{\mathbf{G}}(\mathbf{k},\omega;lz,l'z')$ that relate the field at a plane z in an lth layer with the polarization at a plane z' in an l'th layer. It can be expressed in terms of the functions $\boldsymbol{\mathcal{E}}_q^\nu(\mathbf{k},\omega;z)$ and $\boldsymbol{\mathcal{E}}_q^\nu(-\mathbf{k},\omega;z')$ as [14]

$$\overleftrightarrow{\mathbf{G}}(\mathbf{k},\omega;lz,l'z') = -\frac{4\pi}{k_l^2}\hat{\mathbf{z}}\hat{\mathbf{z}}\delta(z-z')\delta_{ll'} + \sum_{q=p,s}\overleftrightarrow{\mathbf{G}}'_q\,(\mathbf{k},\omega;lz,l'z'),$$

$$\begin{aligned}\overleftrightarrow{\mathbf{G}}'_q\,(\mathbf{k},\omega;lz,l'z') &= \frac{2\pi i}{\beta_n}\frac{\xi_q}{t^q_{0/n}}\left[\boldsymbol{\mathcal{E}}^0_{ql}(\mathbf{k},\omega;z)\boldsymbol{\mathcal{E}}^n_{ql'}(-\mathbf{k},\omega;z')\theta(lz-l'z')\right.\\ &\left.+\boldsymbol{\mathcal{E}}^n_{ql}(\mathbf{k},\omega;z)\boldsymbol{\mathcal{E}}^0_{ql'}(-\mathbf{k},\omega;z')\theta(l'z'-lz)\right],\end{aligned} \tag{7}$$

where $\theta(lz-l'z') = \theta(z-z')$ for $l=l'$ and $\theta(lz-l'z') = \theta(l-l')$ for $l\neq l'$, and $\xi_p = 1$ whereas $\xi_s = -1$. Eqs. (5)-(7), therefore, give the generated field in a simple form in the entire system. Considering the field $\mathbf{E}^{\nu\ \mathrm{out}}$ in the external layers ($\nu = n$ or $\nu = 0$), one finds

$$\begin{aligned}\mathbf{E}^{\nu\ \mathrm{out}}(\mathbf{r},\omega) &= e^{\mathrm{i}\mathbf{K}^\pm_\nu\cdot\mathbf{r}}\,(\frac{\omega}{c})^2\frac{2\pi i}{\beta_\nu}\sum_{q=p,s}\xi_q\hat{\mathbf{e}}^\pm_{q\nu}(\mathbf{k})\\ &\times\sum_l\int_{(l)}\mathrm{d}z'\boldsymbol{\mathcal{E}}^\nu_{ql}(-\mathbf{k},\omega;z')\cdot\chi^{(m)}_l(z'):[\boldsymbol{\mathcal{E}}^{\nu_i}_{q_il}(\mathbf{k}_i,\omega_i;z')E_{q_i}]^m,\end{aligned} \tag{8}$$

where the upper (lower) sign corresponds to $\mathbf{E}^{n\ \mathrm{out}}$ ($\mathbf{E}^{0\ \mathrm{out}}$). The intensity I^ν_q of the q-polarized generated wave in the νth region is given by the corresponding time-averaged Poynting vector. Assuming transparent external layers, this gives

$$I^\nu_q(\mathbf{k},\omega) = (\frac{2\pi}{c})^{m+1}\frac{\omega^2}{[\eta_{\nu_i}(\omega_i)]^m\eta_\nu(\omega)\cos^2\vartheta_\nu}|J^{\nu\nu_1\ldots\nu_m}_{qq_1\ldots q_m}|^2\,[I^{\nu_i}_{q_i}(\mathbf{k}_i,\omega_i)]^m,$$

$$J^{\nu\nu_1\ldots\nu_m}_{qq_1\ldots q_m} = \sum_l \int_{(l)} \mathrm{d}z' \boldsymbol{\mathcal{E}}^{\nu}_{ql}(-\mathbf{k},\omega;z') \cdot \chi^{(m)}_l(z') : [\boldsymbol{\mathcal{E}}^{\nu_i}_{q_i l}(\mathbf{k}_i,\omega_i;z')]^m, \tag{9}$$

with $I^{\nu_i}_{q_i} = (c/2\pi)\eta_{\nu_i}|E_{q_i}|^2$ being the intensity of the ith fundamental beam. In the second factor here, we have used $\beta_\nu = k_\nu \cos\vartheta_\nu$, where $\vartheta_{n(0)} \leq \pi/2$ is defined relative to the positive (negative) z-axis (see Fig. 1) and, together with $\hat{\mathbf{k}} = \cos\varphi\hat{\mathbf{x}} + \sin\varphi\hat{\mathbf{y}}$, describes the direction of the radiation at ω. From $\mathbf{k} = k_\nu(\omega)\sin\vartheta_\nu\hat{\mathbf{k}}$, the angle ϑ_ν is related to the corresponding angles $\vartheta_{\nu_i i}$ of the fundamental waves $[\mathbf{k}_i = k_{\nu_i}(\omega_i)\sin\vartheta_{\nu_i i}\hat{\mathbf{k}}_i]$ through the generalized Snell law

$$k_\nu(\omega)\sin\vartheta_\nu\hat{\mathbf{k}} = \sum_{i=1}^{m} k_{\nu_i}(\omega_i)\sin\vartheta_{\nu_i i}(\pm\hat{\mathbf{k}}_i). \tag{10}$$

Together with Eqs. (3) and (6)-(9), this completes the description of a nonlinear process in a multilayer for all combinations of fundamental- and generated-wave propagation directions and polarizations.

2.4. Cavity effects

In addition to the polarization properties of the process, the effective susceptibility J describes various effects coming from the structural and material properties of the system, e.g., excitation of local guided modes in the system, cavity effects, etc. Focusing attention on cavity effects, we note that, according to Eq. (3), the contribution of each layer to $|J|^2$ includes the function $|t^q_{\nu/l}/D_{ql}|^2$ for each wave, that is, the response (Airy) function of a planar cavity [16]. Letting $r^q_{l\pm} = \xi_q|r^q_{l\pm}|\exp(2i\phi^q_{l\pm})$, this function is transformed into the familiar form generalized, however, for absorption in the system:

$$Y^\nu_{ql}(k) \equiv |\frac{t^q_{\nu/l}}{D_{ql}}|^2 = \frac{|t^q_{\nu/l}|^2}{(1-|r^q_{l-}r^q_{l+}|e^{-\alpha_l d_l})^2 + 4|r^q_{l-}r^q_{l+}|e^{-\alpha_l d_l}\sin^2(\beta'_l d_l + \phi^q_l)}. \tag{11}$$

Here $\alpha_l(k) = 2\beta''_l$ is the absorption coefficient of the cavity and $\phi^q_l(k) = \phi^q_{l+} + \phi^q_{l-}$ the total half phase-shift of the cavity mirrors for a wave. Y^ν_{ql} exhibits a resonance whenever the cavity-mode condition

$$\Phi_{ql}(k) \equiv \beta'_l d_l + \phi^q_l = M_q\pi, \quad M_q = 0, 1, \ldots, \tag{12}$$

is satisfied with the width of the resonance governed by the cavity finesse for the mode [16] $f_{ql}(k) = (\pi/2)\sqrt{F_{ql}}$, where $F_{ql}(k) = 4|r^q_{l-}r^q_{l+}|e^{-\alpha_l d_l}/(1-|r^q_{l-}r^q_{l+}|e^{-\alpha_l d_l})^2$. Thus, any nonlinear layer that supports propagating ($\beta'_l >> \beta''_l$) waves may act as a Fabry-Pérot cavity. Furthermore, according to Eq. (4a), the factor $|t^q_{\nu/l}|^2$ may also exhibit the resonant structure of the form of Eq. (11) with a (nearly) transparent intermediate layer for a wave, so that any layer in the system may, in principle, act as a cavity.

3. Application: Harmonic generation

We illustrate the usefulness of this framework by considering harmonic generation (HG) from a nonlinear slab (l) embedded in a multilayer, as depicted in Fig. 1. We assume a single q'-polarized fundamental wave of the wave vector $\mathbf{k}$ and the frequency ω incident from the top (n) layer at an angle ϑ_n. Adapting the notation appropriately, one can write Eq. (9) as

$$I_q^{(m)\nu} = (\frac{2\pi}{c})^{m+1} \frac{m^2\omega^2}{[\eta_n]^m \eta_\nu^{(m)} \cos^2 \vartheta_\nu^{(m)}} |J_{qq'}^{\nu n}|^2 [I_{q'}^n]^m,$$

$$J_{qq'}^{\nu n} = \int_0^{d_l} \mathrm{d}z' \boldsymbol{\mathcal{E}}_{ql}^{\nu}(-m\mathbf{k}, m\omega; z') \cdot \chi_l^{(m)}(z') : [\boldsymbol{\mathcal{E}}_{q'l}^{n}(\mathbf{k}, \omega; z')]^m, \tag{13}$$

where the superscript (m) denotes quantities that describe the harmonic wave (calculated at mk and/or $m\omega$). With $\nu = n$, this equation describes HG in reflection, and, with $\nu = 0$, HG in transmission. For nondispersive external layers, $\vartheta_n^{(m)} = \vartheta_n$ and $\vartheta_0^{(m)} = \arcsin[(\eta_n/\eta_0^{(m)}) \sin \vartheta_n]$, respectively. Specially, with $\boldsymbol{\mathcal{E}}_{ql}^n(\mathbf{k}, \omega; z) = t_{n/l}^q \hat{\mathbf{e}}_{ql}^- \exp(-i\beta_l z)$ for the bottom layer (l), for a semi-infinite slab one obtains

$$J_{qq'}^{nn}(\infty) = t_{n/l}^{(m)q} [t_{n/l}^{q'}]^m \int_{-\infty}^{0} \mathrm{d}z' e^{-\mathrm{i}(\beta_l^{(m)} + m\beta_l)z'} \xi_q \hat{\mathbf{e}}_{ql}^{(m)+} \cdot \chi_l^{(m)}(z') : [\hat{\mathbf{e}}_{q'l}^-]^m. \tag{14}$$

Of course, as can be easily checked through Eq. (3), this result also emerges directly from Eq. (13) in the limit $d_l \to \infty$. Finally, with $n \to 0$ in Eq. (13), this equation gives HG intensity if the fundamental wave is incident from the bottom (0) layer.

The calculation of $J_{qq'}^{\nu n}$ demands knowledge of the function $\chi_l^{(m)}(z)$. In a phenomenological model, $\chi_l^{(m)}(z)$ for a single-crystal slab is, in general, the sum of the (constant) bulk susceptibility $\chi_{lB}^{(m)}$ and the susceptibilities $\chi_{lS_\pm}^{(m)}$ associated with the slab surfaces. Assuming the surface polarization slightly inside the slab [4] and using Eq. (3), one has

$$J_{qq'}^{\nu n} = \frac{t_{\nu/l}^{(m)q}}{D_{ql}^{(m)}} \left[\frac{t_{n/l}^{q'}}{D_{q'l}} \right]^m \mathcal{J}_{qq'}^{\nu}, \quad \mathcal{J}_{qq'}^{\nu} = \mathcal{J}_{S,qq'}^{\nu} + \mathcal{J}_{B,qq'}^{\nu},$$

$$\begin{aligned} \mathcal{J}_{S,qq'}^{n(0)} \quad = \quad & e^{\mathrm{i}\beta_l^{(m)} d_l} \boldsymbol{\mathcal{E}}_{ql}^{\lessgtr}(-m\mathbf{k}, m\omega; d_l) \cdot \chi_{lS_+}^{(m)} : [e^{\mathrm{i}\beta_l d_l} \boldsymbol{\mathcal{E}}_{q'l}^{<}(\mathbf{k}, \omega; d_l)]^m \\ & + e^{\mathrm{i}\beta_l^{(m)} d_l} \boldsymbol{\mathcal{E}}_{ql}^{\lessgtr}(-m\mathbf{k}, m\omega; 0) \cdot \chi_{lS_-}^{(m)} : [e^{\mathrm{i}\beta_l d_l} \boldsymbol{\mathcal{E}}_{q'l}^{<}(\mathbf{k}, \omega; 0)]^m, \end{aligned}$$

$$\mathcal{J}^{n(0)}_{B,qq'} \quad = \quad \int\limits_0^{d_l} \mathrm{d}z' e^{\mathrm{i}\beta_l^{(m)} d_l} \boldsymbol{\mathcal{E}}^{\lessgtr}_{ql}(-m\mathbf{k}, m\omega; z') \cdot \chi^{(m)}_{lB} : [e^{\mathrm{i}\beta_l d_l} \boldsymbol{\mathcal{E}}^{<}_{q'l}(\mathbf{k}, \omega; z')]^m. \quad (15)$$

With $\boldsymbol{\mathcal{E}}^{\lessgtr}_{ql}(\mathbf{k}, \omega; z)$ from Eq. (3), explicit results for $\mathcal{J}^{\nu}_{S,qq'}$ can be written down immediately for arbitrary m. Performing elementary integrations, one explicitly finds for $\mathcal{J}^{\nu}_{B,qs}$, for example,

$$\mathcal{J}^{n(0)}_{B,ss} = e^{\mathrm{i}(\beta_l^{(m)} + m\beta_l) d_l/2} d_l \left\{ \sum_{t=0}^{m} \frac{m!}{t!(m-t)!} \left(r^s_{l-} e^{\mathrm{i}\beta_l d_l} \right)^t \left[\frac{\sin[\beta_l^{(m)} \pm (m-2t)\beta_l] d_l/2}{[\beta_l^{(m)} \pm (m-2t)\beta_l] d_l/2} \right.\right.$$
$$\left.\left. + r^{(m)s}_{l\mp} e^{\mathrm{i}\beta_l^{(m)} d_l} \frac{\sin[\beta_l^{(m)} \mp (m-2t)\beta_l] d_l/2}{[\beta_l^{(m)} \mp (m-2t)\beta_l] d_l/2} \right] \right\} (-\hat{\mathbf{k}} \times \hat{\mathbf{z}}) \cdot \chi^{(m)}_{lB} : [\hat{\mathbf{k}} \times \hat{\mathbf{z}}]^m, \quad (16a)$$

$$\mathcal{J}^{n(0)}_{B,ps} = e^{\mathrm{i}(\beta_l^{(m)} + m\beta_l) d_l/2} d_l \left[\pm \frac{\beta_l^{(m)}}{k_l^{(m)}} \left\{ r^{(m)s}_{l\mp} \to -r^{(m)p}_{l\mp} \right\} (-\hat{\mathbf{k}}) \cdot \chi^{(m)}_{lB} : [\hat{\mathbf{k}} \times \hat{\mathbf{z}}]^m \right.$$
$$\left. + \frac{(mk)}{k_l^{(m)}} \left\{ r^{(m)s}_{l\mp} \to r^{(m)p}_{l\mp} \right\} \hat{\mathbf{z}} \cdot \chi^{(m)}_{lB} : [\hat{\mathbf{k}} \times \hat{\mathbf{z}}]^m \right], \quad (16b)$$

where $\{ \quad \}$ denotes the expression in the corresponding parenthesis in Eq. (16a). Clearly, $\mathcal{J}^{n(0)}_{B,qp}$ consist of more similar terms with different components of $\chi^{(m)}_{lB}$.

We stress the generality of the above result within the assumptions made. Equations (13)-(15) fully account for all possible reflections of the fundamental and the harmonic wave in an absorbing multilayer. It is worth noticing the appearance of the phase-matching term in $J^{nn}_{qq'}$ (reflection case) and the reinforcement of the corresponding term in $J^{0n}_{qq'}$ (transmission case) as a consequence of wave reflections in the slab [see Eq. (16)]. HG from a semi-infinite medium (the $n-l$ system) or a three-layer ($n-l-0$) system, frequently considered in the literature [2–6,11–13] is described by these equations with the use of the corresponding single-interface Fresnel coefficients and for appropriate m. Finally, when summing $J^{\nu n}_{qq'}$ for different layers, Eq. (13) gives HG intensity from an arbitrary nonlinear multilayered system.

Application of the above result in spectroscopy of multilayers is obvious. Cavity effects in the present case are described by the product $Y^{(m)\nu}_{ql}[Y^{n}_{q'l}]^m$ of the Airy functions and, as mentioned, by a similar form arising in the product $|t^{(m)q}_{\nu/l}|^2[|t^{q'}_{n/l}|^2]^m$ for any intermediate layer. Thus, looking, for example, at the variation of HG intensity with increasing slab thickness, one obtains a rather simple pattern with maxima appearing whenever d_l matches the cavity-mode condition, Eq. (12), either for the fundamental or for the harmonic wave. In the first case, an enhanced HG intensity is given by

$$|J_{qq'}^{\nu n}|^2 = \frac{G_{\nu/l}^{(m)q}[G_{n/l}^{q'}]^m}{1 + F_{ql}^{(m)} \sin^2 \Phi_{ql}^{(m)}} |\mathcal{J}_{qq'}^{\nu}|^2, \quad G_{\nu/l}^q = \frac{|t_{\nu/l}^q|^2}{(1 - |r_{l-}^q r_{l+}^q| e^{-\alpha_l d_l})^2}, \tag{17}$$

with $\beta_l d_l = M_{q'}\pi - \phi_l^{q'} + i(\alpha_l/2)d_l$ in $\mathcal{J}_{qq'}^{\nu}$. A similar result is obtained for the harmonic wave coinciding with a cavity mode. In this case, however, HG intensity should normally be smaller since $Y^{(m)}$ rather than Y^m is at a peak value. Of course, it may happen that either because of the weak reflection or, more likely, because of the strong absorption of a wave, one of the Airy functions becomes ineffective. In this case, one observes variations of HG intensity governed by the parameters of one beam only. All these characteristic cavity effects have been clearly observed in SHG from supported thin films [2–6].

In quantum optics, the system considered represents a nonlinear cavity formed by the two multilayered mirrors. Solid-state planar cavities of this form have recently been employed as parts of various surface-emitting devices, e.g., a nonlinear planar cavity may be integrated with a semiconductor surface-emitting laser to form a monolithic surface-emitting harmonic generator [8–10]. When specified to the $k = 0$ case ($r_{l\pm}^q = \xi_q r_{l\pm}$), this theory provides the theoretical background for such a device. Thus, with the condition $\eta_l d_l = (M - \phi_l/\pi)\lambda/2$, Eqs. (13) and (17) directly give the power conversion efficiency ($\sim I_q^{\nu}/[I_{q'}^n]^m$) of a nonlinear cavity designed to resonate with the fundamental wave. Using Eq. (4a), the enhancement factors $G_{\nu/l}^q$ are rewritten in the terminology of quantum optics as

$$G_{\nu/l}^q = \frac{\eta_\nu \eta_l}{|n_l|^2} \frac{T_\pm}{(1 - \sqrt{R_- R_+} e^{-\alpha_l d_l})^2}, \tag{18}$$

where $R_\pm = |r_{l\pm}|^2$ and $T_\pm = (\eta_\nu/\eta_l)|t_{l/\nu}|^2$ is the reflectivity and transmissivity (from the cavity side), respectively, of the upper (lower) cavity mirror. According to Eqs. (3) and (15), the overlap integrals $\mathcal{J}_{qq'}^{\nu}$ become

$$\mathcal{J}_{qq'}^{n(0)} = \int_0^{d_l} \mathrm{d}z' \chi(z') f_\mp^{(m)}(z'_\mp)[f_-(z')]^m, \quad f_\mp(z) = e^{\mathrm{i}k_l d_l} \left[e^{-\mathrm{i}k_l z} - r_{l\mp} e^{\mathrm{i}k_l z} \right], \tag{19}$$

where $\chi = (-\hat{\mathbf{q}}) \cdot \chi_l^{(m)} : [\hat{\mathbf{q}}']^m$, with $\hat{\mathbf{q}}$ describing the polarization of a wave that propagates in the z direction. For highly-reflecting ($R_\pm \simeq 1$) cavity mirrors, the approximation $f_\mp(z) \simeq -2i \exp[i(k_l d_l + \phi_{l\mp})] \sin(k_l z + \phi_{l\mp})$ is appropriate. Therefore, for a structured cavity described by an effective dielectric function ε_l, Eq. (19) provides a simple recipe for quasi–phase–matching [17] in HG, which is the method usually utilized for maximizing $\mathcal{J}_{qq'}^{\nu}$ in semiconductor-based nonlinear cavities [7–10]. The conversion efficiency of a cavity designed to resonate with the harmonic wave $[\eta_l^{(m)} d_l = (M^{(m)} - \phi_l^{(m)}/\pi)\lambda^{(m)}/2]$, or with both the harmonic and the fundamental wave simultaneously, is given by Eqs. (13) and (17)-(19), with an obvious modification of the denominator in Eq. (17).

4. *Summary*

In summary, we have developed a transparent general formalism for consideration of nonlinear processes in layered systems. In this work we have specially emphasized its usefulness in discussing cavity effects that may be observed or exploited in nonlinear spectroscopy of multilayers and quantum optics. Therefore, attention has been paid to processes with unbound waves in a system. It should be noted, however, that this theory also applies to nonlinear processes with guided modes in a system mediated, e.g., by a prism coupler. To consider such a process, one of the external layers in Fig. 1 should be regarded as the prism and its effect can be easily analyzed using the properties of the generalized Fresnel coefficients.

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NELINEARNA GENERACIJA SVJETLA U VIŠESLOJNIM SISTEMIMA: EFEKTI REZONATORA

Koristeći nedavno izvedeni izraz za Greenovu funkciju slojevitih sistema razvijen je sažet i jasan postupak za proračun nelinearnih procesa u višeslojnim strukturama. Teorija je posebno prikladna za analizu efekta rezonatora (interferencije višestruko reflektiranih valova) na nelinearnu generaciju svjetla u višeslojnim sistemima i planarnim rezonatorima. To je ilustrirano razmatranjem procesa harmonijske generacije svjetla u strukturama s jednim nelinearnim slojem te kratkom diskusijom efekta rezonatora u takvim sistemima.