Design and theoretical analysis of resonant cavity for second-harmonic generation with high efficiency

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An improved theoretical model is built to discuss the second-harmonic generation (SHG) with high efficiency in a microcavity. Meanwhile, the configuration of the cavity is optimized according to it. It is found that when the length of the cavity leads to a π phase mismatch between fundamental and second-harmonic wave, the cavity reaches its optimum working condition. The SHG efficiency has been calculated for a 10.6 μm laser in a 106.3 μm ⟨111⟩ GaAs cavity. © *2011 American Institute of Physics*. [doi:10.1063/1.3544056]

Second-harmonic generation (SHG) is always a focus because of the applications in coherent green and blue lasers, high density information storage, ophthalmology, etc.[1] Lots of nonbirefringent materials, such as semiconductors[2] and organic molecules,[3] have large second-order susceptibility. However, it is still a technological challenge to realize phase matching or quasi-phase-matching (QPM) in most of the existing materials.

To overcome these limitations and to enhance the conversion efficiency, the development of non-phase-matching method has been reported.[4,5] In 1966, Ashkin *et al.* pointed out that a cavity could improve the SHG efficiency[6] due to the enhancement of fundamental wave (FW) by resonance, which is proved by experiments.[7,8] In their calculation, the concrete nonlinear process is simplified as a loss of FW to satisfy the energy conservation. Though ignoring details of SHG, it fitted the experiment well.[9] In 1995, Rosencher *et al.* built an approximate theory for SHG in *Fabry-Pérot cavity*, considering the specific processes,[10] but it is only suitable for low pump power condition and violates energy conservation when high conversion efficiency is achieved. This is because the amplitude of the FW in the cavity is calculated separately without considering the energy conversion from FW to the second-harmonic wave (SHW).

In order to discuss the high efficiency scenario, a more accurate theory is needed, in which both the concrete process and energy conservation are contained. Here, we considered a resonant cavity for FW, in which the SHW resonates weakly or even passes the working media only twice. This assumption leads to a relatively weak SHW compared to FW, so that an undepleted-pump approximation can be applied,[11] assuming a uniform amplitude of the FW of the cavity.[12] To guarantee the energy conversion, one can simply impose it as a condition by letting the energy output from the cavity equal to the input.

As demonstrated in Fig. 1, the cavity for SHG is a piece of nonlinear media M with dielectric mirrors F_1 and F_2 on both parallel sides. The length L of M meets the resonance condition for FW. The refractive index of M is n_F for FW and n_S for SHW. The FW is normally incident from the F_1; the SHW is emitted from F_2. The reflectance and transmittance of F_1 for FW are R_F and T_F, respectively. The reflectance of F_1 for the SHW should be near 100% to avoid SHW output from F_1. Meanwhile, the reflectance of F_2 for FW should be near 100% to avoid leak of FW from F_2 and for SHW the transmittance and reflectance of F_2 are T_S and R_S, respectively. R_S should be small to promise the weak resonance of SHW.

As an optimal case, the efficiency of the cavity is assumed to be 100%. The amplitude of the incident FW and that of the reflected FW from F_1 are A_1 and $\sqrt{R_F}A_1$, respectively. The amplitude of FW transmitted the medium and reflected by F_2 is $-\sqrt{T_F}\sqrt{n_F/n_1}A_F$, where A_F is the amplitude of FW in the cavity. (The minus sign results from the π phase shift, which is chosen for convenience without losing generality.) Because of the resonance of FW, A_1 and A_F are both real numbers. Once FW is incident into the cavity, neither transmission nor reflection is allowed. To avoid reflection, the destructive interference between the FW reflected by F_1 and the one reflected by F_2 is utilized, as described in Eq. (1),

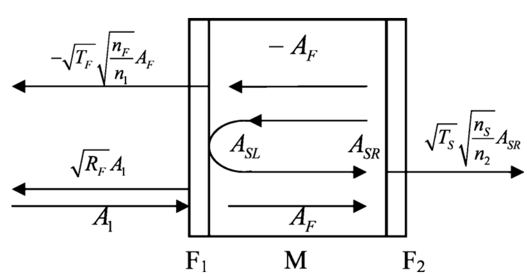

FIG. 1. The schematic structure of the resonance cavity for SHG with high efficiency. (F_1 and F_2 are optical dielectric mirrors; M is the working media; A_1 and A_2 stand for the amplitudes of the FW and SHW out of the cavity, respectively; A_F means the amplitude of the FW; and A_{LS} and A_{RS} are the amplitudes of SHW at the interface of F_1 and M and the interface of M and F_2, respectively).

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$$\sqrt{T_F}\sqrt{n_F/n_1}A_F - \sqrt{R_F}A_1 = 0. \tag{1}$$

The conservation of energy is expressed in Eq. (2). A_{SR} is the amplitude of SHW at the interface of M and F_2. Here,

$$2\varepsilon_0 c n_1 |A_1|^2 = 2\varepsilon_0 c n_2 |\sqrt{T_S}\sqrt{n_S/n_2}A_{SR}|^2. \tag{2}$$

The amplitude of the SHW can be calculated via Eq. (3); E_S is the electric field intensity of the SHW and d_{eff} is the effective value of second-order susceptibility. Here,

$$\nabla^2 E_S - \frac{n_S^2}{c^2}\frac{\partial^2 E_S}{\partial t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}[2\varepsilon_0 d_{eff}(E_F)^2]. \tag{3}$$

Considering the standing wave condition for FW, the solution can be expressed as Eq. (4),

$$E_S = \alpha A_F^2\{\exp[i(2k_F z - 2\omega t)] + \exp[i(-2k_F z - 2\omega t)]\} + A_{S1}\exp[i(k_S z - 2\omega t)] + A_{S2}\exp[i(-k_S z - 2\omega t)], \tag{4}$$

where

$$\alpha = 2d_{eff}/[(n_F)^2 - (n_S)^2]. \tag{5}$$

The boundary condition for SHW at the interface of F_1 and M is shown in Eq. (6a), and the interface of F_2 and M is described by Eq. (6b). The minus signs also come from the π phase shift. Here,

$$A_{S1} + \alpha A_F^2 = -(A_{S2} + \alpha A_F^2), \tag{6a}$$

$$A_{S2}e^{-ik_S L} + \alpha A_F^2 e^{-i2k_F L} = -\sqrt{R_S}(A_{S1}e^{ik_S L} + \alpha A_F^2 e^{i2k_F L}). \tag{6b}$$

Because of the standing wave condition for FW, each phase item can be multiplied by $\exp[i2nk_F\cdot L](=1)$.

Combining Eqs. (1), (2), and (6), one obtains Eqs. (7),

$$\sqrt{\frac{R_F}{T_F}} = \left(\frac{1}{|A_1|}\frac{T_F n_F}{R_F\sqrt{n_1 T_S n_S}}\cdot\frac{1}{\alpha}\cdot\left|\frac{e^{i2\Delta kL} - \sqrt{R_S}}{e^{i2\Delta kL} - 2e^{i\Delta kL} + 1}\right|\right)^{1/2},$$

$$|A_{SR}| = \frac{T_F n_F}{R_F T_S n_S}\cdot\frac{1}{\alpha}\cdot\left|\frac{e^{i2\Delta kL} - \sqrt{R_S}}{e^{i2\Delta kL} - 2e^{i\Delta kL} + 1}\right|. \tag{7}$$

Here, $\Delta k = 2k_F - k_S$ and $|A_{SR}|$ is the amplitude of the SHW at the right interface. The result indicates that the higher second-order susceptibility results in weaker FW to achieve high efficiency.

Now it is necessary to discuss the amplitudes of FW and SHW because of the limitation imposed by the laser damage threshold.

Because A_F is proportional to $\sqrt{R_F/T_F}$ when A_1 is a constant, it is enough to discuss the value of $\sqrt{R_F/T_F}$ as the first equation in Eq. (7) shows, and the picture is shown in Fig. 2. A_F reaches its minimum when $\Delta k\mathrm{L} = (2m+1)\pi$ (m is an integer). This is because the SHW travels a length of L in the cavity with phase mismatch of $(2m+1)\pi$; reflected by F_1 with a π shift, it will travel a length of L again. This process is very similar to that of QPM in superlattices and the only difference is the direction of the electric field of SHW alternates rather than the orientation of nonlinear susceptibility. Meanwhile, larger R_S leads to weaker A_F under the double resonance with $(2m+1)\pi$ phase mismatching. Yet, large R_S will lead to the collapse of the above approximation.

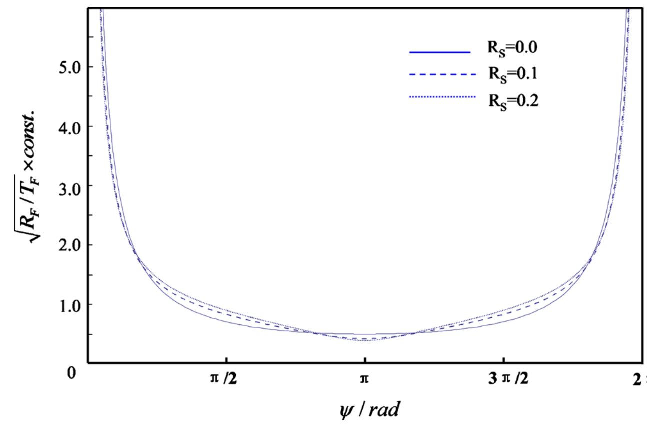

FIG. 2. (Color online) The map of $\sqrt{R_F/T_F}$ as a function of R_S and $\Delta k\mathrm{L}(\psi)$. (R_F and T_F are the reflectance and transmittance of F_1, R_S is the reflectance of F_2, L is the length of the cavity, and Δk is the wave vector mismatch).

Equation (2) shows that when A_1 is constant, the smaller the value of R_S, the weaker A_{SR} will be. Hence, as R_S increases, A_F and A_{SR} exhibit different trends.

When $c = 2m\pi$, the value of Eq. (7) diverges, although this is also a double resonance model. This is because the phase mismatch equates to $2m\pi$ and the amplitude of SHW is equal to zero besides the interface of M and F_2.

One may be confused by the absence of boundary condition for FW. In a strict theory, the energy conversion should be met automatically with right boundary conditions. Yet, they are not compatible with each other under undepleted-pump approximation. Considering strict boundary condition, one gets the result of Rosencher. When one loosens the boundary conditions, two things should be kept in mind. One is that the amplitude of the FW is determined by the energy conversion equation; the other is that the SHW wave is relatively weak, so that it does not influence much the phase of the FW, which is very close to the phase in a cavity without the nonlinear process (as will be discussed later).

The above discussion assumes that the power and wavelength of FW equal to the theoretical values and promise 100% efficiency. A more general case in which the wavelength and amplitude of the incident FW deviate from the ideal value should be considered.

For convenience, all the symbols below are generated by adding an apostrophe on their counterparts without changing meanings.

Then FW in the cavity is

$$E_F' = |A_F'|\exp[i(k_F'\cdot x + \theta)] - |A_F'|\exp[-i(k_F'\cdot x - \theta - 2\varphi)]. \tag{8}$$

Here, $\varphi = k_F'\cdot L$ and $\theta = \arctan\{\sqrt{R_F}\cdot\sin(2\varphi)/[1 - \sqrt{R_F}\cdot\cos(2\varphi)]\}$. As mentioned above, all these phase parameters of FW are determined by a Fabry-Pérot cavity without considering nonlinear interactions.

The FW reflected by the cavity A_r' is described by Eq. (9),

$$A_r' = \sqrt{R_F}|A_1'| - \sqrt{T_F}\sqrt{n_F/n_1}|A_F'| \times \exp[i(\theta + 2\varphi)]. \tag{9}$$

Repeating the similar process of Eqs. (3)–(5), (6a), and (6b), the amplitude of the SHW is expressed by Eq. (10). Here, R_S is set to be 0. Here,

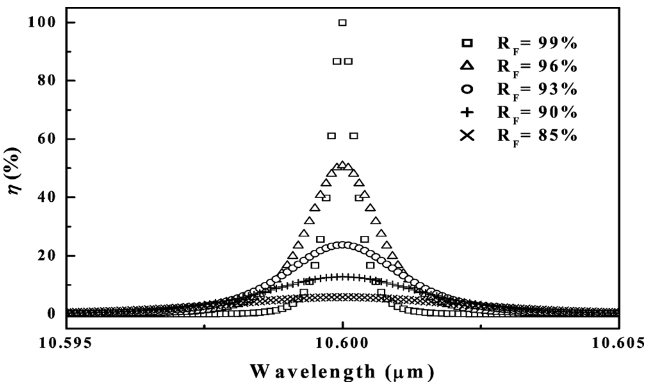

FIG. 3. Calculated SHG efficiency for 106.3 μm GaAs cavity with different R_F (R_F is the reflectance of mirror F_1). The pump power is 7.5 MW/cm^2.

$$|A'_{SR}| = \alpha|A'_F|^2 \times |e^{i(4\varphi-\Delta k'\cdot L)} - 1 - e^{i4\varphi} + e^{i\Delta k'\cdot L}|. \qquad (10)$$

Conservation of energy can be expressed by Eq. (11),

$$2\varepsilon_0 cn_1|A'_1|^2 = 2\varepsilon_0 cn_S|A'_{SR}|^2 + 2\varepsilon_0 cn_1|A'_r|^2. \qquad (11)$$

From Eqs. (9)–(11), the efficiency η is described as Eq. (12),[13]

$$\eta + R_F \times \left(1 - 2\frac{\sqrt[4]{\eta}}{\sqrt{\zeta \times \Xi}}\cos(2\varphi + \theta) + \frac{\sqrt{\eta}}{\zeta \times \Xi}\right) = 1, \qquad (12)$$

where $\Xi=(e^{i(4\varphi-\Delta k'\cdot L)}-1-e^{i4\varphi}+e^{i\Delta k'\cdot L})/(1-2e^{-i\Delta k\cdot L}+e^{-i2\Delta k\cdot L})$ and $\zeta=|A'_1/A_1|$.

Now, let us take $\langle 111\rangle$ GaAs (n_F=3.2919, n_S=3.3168, and $d_{eff}=10^{-10}$ m/V[12]) as working medium to calculate the parameters of the cavity for a 10.6 μm laser. The length of the cavity equals $p\lambda_F/2n_F$, where p is the number of FW antinodes in it. In order to select a working point close to the near QPM region, p is set to be 66, corresponding to a length of 106.3 μm.

Theoretically, this cavity can convert a 7.5 MW/cm^2 FW into SHW with high efficiency when R_F=0.99 and R_S=0. In experiment, the bandwidth of pump must be considered. Then the efficiency of the 106.3 μm GaAs cavity as a function of R_F and wavelength of a 7.5 MW/cm^2 laser is calculated and demonstrated in Fig. 3, which behavior is similar to the numerical results of Klemens and Feinman,[14] although the boundary conditions are not exactly the same. When considering the bandwidth of the ultrafast laser, the efficiency can be estimated as 75% for 1 ns pulse, 9.5% for 100 ps, 1.0% for 10 ps, and 0.13% for 1 ps.

Other effects, such as the third-harmonic generation, the sum frequency of $\omega+\Delta\omega$ and $\omega-\Delta\omega$ waves, the intensity-dependent refractive index, etc., which may also affect the efficiency of the cavity, are not fully considered here.

Now, let us review the boundary condition for FW. According to the above calculation, A_F has the order of 10^7 V/m, while α is in the order of 10^{-9} m/V, so A_S has an order of 10^5 V/m. The feedback from the SHW to FW has the order of 10^3 V/m, which is too small to affect the phase of FW at the boundary.

In conclusion, improvements have been made to the structure of the cavity for SHG; especially the properties of the dielectric mirrors for effective SHG are discussed. A more accurate model is provided to determine the parameters, the intensities of FW and SHW. This model also illustrates the variations of the amplitude of FW and SHW as functions of the parameters of the cavity and so provides a guideline to determine the best working point. Finally, GaAs is taken as an example to determine the necessary parameters and the efficiency.

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