Second harmonic generation in disordered media: Random resonators

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We theoretically study the effect of random resonators on the conversion efficiency of fundamental mode propagating through disordered nonlinear dielectric film. Only resonators with double-resonant properties, i.e., which can trap both the fundamental and second harmonic modes, contribute to local generation of the second harmonic light of high intensity. We calculate the density of such resonators. The parameters of the random media under which all the random resonators with the given quality factors have the double-resonant properties are found.

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I. INTRODUCTION

Generation of a second harmonic mode due to nonlinear properties of optical media has been the subject of intensive research during the last decades.[1] The main problem in realization of an efficient conversion of the fundamental mode into the second harmonic mode is that the modes should satisfy the phase matching condition. Due to the frequency dependence of the dielectric constant there is a phase mismatch for light propagating through nonlinear homogeneous media. The phase mismatch introduces a limitation on the second-harmonic yield. There are different methods to overcome the phase mismatch in the second harmonic generation process. One of them is to employ the photonic crystals, in which the photon dispersion can be tailored in any predictable way.[2] Therefore at some photonic frequencies the quasi-phase-matching conditions can be realized in photonic crystals.[3] Another efficient method to enhance the generation of second harmonic mode is to incorporate the nonlinear media in a resonant cavity.[4] In this case the efficient frequency conversion is achieved if both the fundamental mode and the second harmonic mode are the resonant modes of the cavity. Therefore the cavity has a double-resonant property. If the fundamental mode propagates through such a cavity then the high intensity second harmonic mode is generated.

The double resonant cavity can be realized as a localized mode of a photon. The localization can be easily achieved in photonic periodic structures with complete bandgaps. Adding the defect to such system produces the localized modes of the light within the bandgap of photonic crystal.[5–7] The parameters of the defect should be tailored so that the light at both the fundamental frequency and the double frequency is localized.

The localization of light can also be achieved in strongly disordered media. Here the light should be localized at the fundamental and second harmonic frequencies within the same spatial region. In many experimental realization of dielectric disordered media the disorder is not strong enough to localize the light. But even in this case we have resonators, which are called random resonators.[8–10] Such random resonators do not localize but trap the light for a very long time. The analog of such random resonators in disordered electronic systems are prelocalized electronic states.[11]

The random resonators manifest themselves as the bright spots in the spatial distribution of emission intensity on the surface of disordered dielectric films, i.e., in the speckle pattern. In disordered media with amplification the random resonators can result in coherent random lasing.[12–16] Although random resonators are sparse, the intensity of light within such resonators are very large due to large quality factors of such resonators.[8–10]

In the present paper we show that random resonators can be not only the source of coherent random lasing but also enhance the generation of the second harmonic mode in nonlinear disordered media. Namely, we discuss the distribution of the intensity of the second harmonic mode within disordered system with the emphasis on very large intensities. We show that the large intensity of the second harmonic mode is realized within the random resonators with the double resonant properties, i.e., random resonators can trap both the fundamental and the second harmonic modes. In addition to the double resonant properties the random resonators should satisfy the phase matching condition, i.e., the second harmonic mode should have twice the angular momentum of the fundamental mode. Finally, below we reformulate the problem of finding the distribution of the intensity of the second harmonic mode in terms of the problem of finding the distribution of random resonators with a given trapping time and which satisfy the double resonant property. Below we consider only the two-dimensional case, i.e., disordered film.

The net enhancement of the intensity of second harmonic mode in strongly disordered media has been observed experimentally[17] in porous gallium phosphate structure. The results of Ref. 17 illustrate an importance of disorder in the generation of the second harmonic mode in disordered media. We want to emphasize the difference between Ref. 17 and our study. In the present paper we address the problem of not the average intensity of the second harmonic mode but the distribution of the very large intensity of the second harmonic mode within disordered media. This means that if we are looking at the spatial distribution of the second harmonic mode on the surface of the sample, and we study the density of very bright spots at the second harmonic frequency.

The paper is organized as follows. In Sec. II we describe the structure of random resonators and the trapping modes within such resonators. We discuss the relevance of random resonators for local generation of the second harmonic modes in nonlinear media. In Sec. III we introduce the main system of equations to find the density of random resonators, which can trap both the fundamental and the second har-

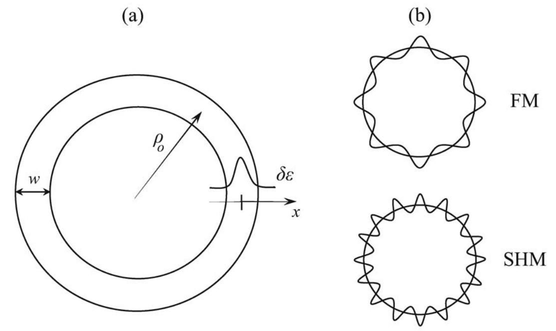

FIG. 1. (a) The structure of random resonator in disordered dielectric media is shown schematically. The resonator is described by small enhancement of dielectric constant within a ring of radius ρ_0. The width of the ring is much smaller than its radius. (b) Illustration of the fundamental mode (FM) and the second harmonic mode (SHM) trapped within the random resonator. The phase matching condition for fundamental and the second harmonic modes determine the relation between their angular momenta: the angular momentum of the second harmonic mode is twice the angular momentum of the fundamental mode.

monic modes. In Sec. IV we present the numerical solution of the main system of equations and discuss the results.

II. RANDOM RESONATORS AND SECOND HARMONIC GENERATION

The random resonator appears in weakly disordered media and plays the role of a trap for a propagating light. The random resonator has a shape of a ring, within the ring dielectric constant is enhanced by some small value $\delta\varepsilon$. The fluctuation $\delta\varepsilon(r)$ is azimuthally symmetric and nonzero only within narrow interval w within the ring.[8] Such a configuration plays the role of the waveguide for light and is shown schematically in Fig. 1. The trapping time of the light within the random resonator is determined by evanescent leakage, which depends on the curvature of the waveguide. The waveguide with the smallest evanescent leakage should have the same curvature at all points, therefore the resonator should have the shape of the ring.

Due to azimuthal symmetry of the system the modes of the light trapped within the ring are characterized by an angular momentum m which is equal to the number of the wavelength along the ring, see Fig. 1(b). The radius of the ring is related to the angular momentum through the relationship $\rho_0 = m/\varepsilon^{1/2}k_0$, where k_0 is the wave vector and ε is the average dielectric constant of the medium. The random resonator should satisfy the condition that it can trap the light for a given trapping time $\mathcal{T}$. It means that the resonator is characterized by a given quality factor, the Q factor, which is related to the $\mathcal{T}$ and frequency ω through $Q \approx \omega\mathcal{T}$.

The probability or the density of random resonators is exponentially small and is not universal. This means that it depends not only on the mean free path of the wave within disordered media but also on other parameters of disorder, such as correlation radius. The density of random resonators with a given quality factor Q has the following expression:

$$\mathcal{N} = \mathcal{N}_0 e^{-S(kl,Q,kR_c)}, \tag{1}$$

where $\mathcal{N}_0$ is a prefactor,[10,21] l is the transport mean free path of the wave, and R_c is the correlation radius of disorder.

Within the optimal fluctuation approach the exponential factor in Eq. (1) can be calculated. The main steps in these calculations are the following.[8] In the scalar wave model the waveguide modes of the random resonator at frequency ω have the general form

$$\Psi(\rho,\theta) = (2\pi)^{-1/2}\psi(\rho)\exp(im\theta), \tag{2}$$

where $i=\sqrt{-1}$ and ρ, θ are polar coordinates. With this expression the scalar wave equation for the trapped mode becomes

$$\nabla_\rho^2\psi + k_0^2\delta\varepsilon(r)\psi = -k_0^2\epsilon\psi, \tag{3}$$

where $\epsilon = (m/k_0\rho_0)^2 - \varepsilon$ describes the "eigenvalue" of the mode and can be related to the quality factor of random resonator $\epsilon = \varepsilon(3\ln Q/2m)^{2/3}$. This relation is based on the expression for the tunneling of a light through the trapping barrier.[8]

Then the last step in these calculations is that we need to find the most probable fluctuation of dielectric constant $\delta\varepsilon(r)$ for which the resonator mode satisfies the wave equation (3), i.e., the mode has a given quality factor Q. To define the most probable fluctuation we assume that disorder has a Gaussian form, i.e., the probability W of realization of fluctuation $\delta\varepsilon(r)$ is determined by the following expression:

$$\ln W = -\frac{1}{2\Lambda^2}\int\int d\mathbf{r}d\mathbf{r}'\,\delta\varepsilon(\mathbf{r})\delta\varepsilon(\mathbf{r}')\kappa(\mathbf{r}-\mathbf{r}'), \tag{4}$$

where Λ is the r.m.s. of the disorder, and the kernel $\kappa(\mathbf{r}-\mathbf{r}')$ is the inverse of the correlator $K(\mathbf{r}-\mathbf{r}')$, defined as

$$\langle\delta\varepsilon(\mathbf{r})\delta\varepsilon(\mathbf{r}')\rangle = \Lambda^2 K(\mathbf{r}-\mathbf{r}'). \tag{5}$$

Therefore the problem of finding the density of random resonators is reformulated in terms of the problem of finding the azimuthally symmetric fluctuation of dielectric constant, which provides the largest value of probability W and satisfies Eq. (3).

The quality factor of corresponding random resonator is large, $Q \approx \omega\mathcal{T} \gg 1$, which means that for the mode, trapped within the resonator, electric field is enhanced in $\omega\mathcal{T}$ times compared to the average value. Now we are going to discuss the relation between the properties of random resonators and generation of the second harmonic mode in disordered media. The wave equations, which describe the properties of light at the fundamental and the second harmonic frequencies in the disordered media with dielectric constant $\varepsilon(r)$, have the following form:

$$\nabla^2\Psi_1 + k_0^2\varepsilon(r)\Psi_1 = 0, \tag{6}$$

$$\nabla^2\Psi_2 + 4k_0^2\varepsilon(r)\Psi_2 = -16\chi\pi k_0^2\Psi_1^2, \tag{7}$$

where χ is a nonlinear coefficient, and Ψ_1 and Ψ_2 are the wave fields, corresponding to fundamental and the second harmonic modes, respectively. In Eq. (7) we have an additional term proportional to Ψ_1^2 due to nonlinear properties of the media.[18] Here we disregard the effects of the second harmonic mode on the fundamental mode.

As we already mentioned in the Introduction we study not the average intensity of the second harmonic mode but the distribution of its large intensity. The large intensity of the second harmonic mode is realized only at some spatially localized spots. The density of such spots is exponentially small, therefore we can disregard the "interaction" between the second harmonic modes in different spots. The intensity of the second harmonic mode within the spot is large. The accumulation of large intensity can be done by two different ways.

(i) The second harmonic mode is trapped by the spot, but generated outside of the spot. Since the most efficient trapping is realized by the random resonators then we have the trapping of the second harmonic mode by random resonator. In this case the enhancement of the second harmonic field within the random resonator compared to the average value is $\sim\mathcal{T}_2$, where $\mathcal{T}_2$ is the trapping time of the second harmonic mode. We assume that the average fundamental field, i.e., the field outside of random resonator, is $\Psi_{1,0}$. Then the average second harmonic field generated by $\Psi_{1,0}$ is $\Psi_{2,0}\sim\chi\Psi_{1,0}^2$. This is the second harmonic field outside of the random resonator. Since the field inside of the random resonator is enhanced by coefficient $\mathcal{T}_2$, we can find the second harmonic field inside of the resonator

$$\Psi_2 \sim \mathcal{T}_2\Psi_{2,0} \sim \chi\mathcal{T}_2\Psi_{1,0}^2. \tag{8}$$

Therefore, in this case the second harmonic mode is generated outside of the random resonator and the enhancement of the intensity is due to the trapping of the second harmonic mode only.

(ii) In the second case the second harmonic mode is generated within the spot. This means that there is a large intensity of the fundamental mode within the spot, which generates the second harmonic mode. The large intensity of the fundamental mode can be achieved only if the mode is trapped by the spot. Therefore, the spot is again a random resonator. But now the random resonator is trapping the fundamental mode. At the same time it is known that, in general, the generation of the second harmonic mode within the resonator is efficient only if both modes, fundamental and the second harmonic, are trapped by the same resonator. Therefore the condition of efficient generation of the second harmonic mode within random resonator is that the random resonator is the trap for fundamental and the second harmonic modes, i.e., the resonator has the double-resonant property. If the average fundamental field outside of the random resonator is $\Psi_{1,0}$ then the field inside of the resonator is $\Psi_1\sim\mathcal{T}_1\Psi_{1,0}$. This is because the enhancement of the fundamental field by random resonator is $\mathcal{T}_1$, where $\mathcal{T}_1$ is the trapping time of the fundamental mode. Then we can estimate the enhancement of the second harmonic field within the random resonator as

$$\Psi_2 \sim \chi f_m\mathcal{T}_2\Psi_1^2 \sim \chi f_m\mathcal{T}_2\mathcal{T}_1^2\Psi_{1,0}^2. \tag{9}$$

The coefficient f_m takes into account the matching condition. The matching condition of the fundamental and the second harmonic modes can be reformulated in terms of the angular momentum. Namely, due to azimuthal symmetry, the fields have the following form:

$$\Psi_1(\rho,\theta) = (2\pi)^{-1/2}\psi_1(\rho)\exp(im\theta), \tag{10}$$

$$\Psi_2(\rho,\theta) = (2\pi)^{-1/2}\psi_2(\rho)\exp(im_2\theta), \tag{11}$$

where m_2 is the angular momentum of the second harmonic mode. Since the right-hand side of Eq. (7) contains $\Psi_1^2\propto\exp(2im\theta)$, then Ψ_2 should have the same angular dependence as Ψ_1^2. This means that the angular momentum of the second harmonic mode is equal to $m_2=2m$. This is the phase matching condition for the trapped fundamental and the second harmonic modes. Under this condition the factor f_m is maximum.

Therefore when the matching conditions are satisfied the second harmonic field in the double-resonant random resonator is

$$\Psi_2 \sim \chi\mathcal{T}_2\mathcal{T}_1^2\Psi_{1,0}^2. \tag{12}$$

We can see that the final result is proportional to the trapping time of the second harmonic mode $\mathcal{T}_2$. Therefore the trapping time $\mathcal{T}_2$ should be large, which confirms our statement that both modes should be trapped within the same random resonator.

Comparing two cases (i) [Eq. (8)] and (ii) [Eq. (12)] of generation of local field at the second harmonic frequency we conclude that the second case is the most efficient one. The second harmonic field in the second case is $\mathcal{T}_1^2(\mathcal{T}_1^2\gg 1)$ times larger than in the first case. This means that the second harmonic mode of the highest intensity is generated within the random resonator with the double resonant property, i.e., the random resonator can trap both the fundamental and the second harmonic mode and the angular momentum of the second harmonic mode is twice the angular momentum of the fundamental mode. The intensity of the second harmonic mode within such resonator is

$$I_2 \sim \chi^2\mathcal{T}_2^2\mathcal{T}_1^4. \tag{13}$$

Since the intensity I_2 is proportional to $\mathcal{T}_1^4$ we characterize the random resonators by the trapping time of the fundamental mode, i.e., below we find the distribution of the trapping time $\mathcal{T}_1$ or quality factor Q_1 of the random resonators with the double resonant property.

III. THE MAIN SYSTEM OF EQUATIONS: OPTIMAL FLUCTUATION APPROACH

To find the distribution function of the intensity of second harmonic mode we calculate the density (probability) of random resonators with the double resonant property. The calculations are the same as for the usual random resonators, but now we have one more condition. Namely, the resonator should also trap the second harmonic mode. To find the equations, which describe the resonator modes, we substitute $\varepsilon(r)=\varepsilon(\omega)+\delta\varepsilon$ and Eqs. (10) and (11) into the wave equations (6) and (7). Taking into account that $m_2=2m$ and the fluctuations of dielectric constant $\delta\varepsilon(\rho)$ are azimuthally symmetric, we obtain the following equations:

$$\nabla_\rho^2 \psi_1 + k_0^2 \delta\varepsilon(\rho) \psi_1 = -k_0^2 \epsilon_1 \psi_1, \quad (14)$$

$$\nabla_\rho^2 \psi_2 + 4k_0^2 \delta\varepsilon(\rho) \psi_2 = -4k_0^2 \epsilon_2 \psi_2 - 16\chi\pi k_0^2 \psi_1^2, \quad (15)$$

where $\epsilon_1 = (m/k_0\rho_0)^2 - \varepsilon_1$, $\epsilon_2 = (m/k_0\rho_0)^2 - \varepsilon_2$ and $\varepsilon_1 = \varepsilon(\omega)$, $\varepsilon_2 = \varepsilon(2\omega)$ are dielectric constants at the fundamental and second harmonic frequencies. Here we assume that fluctuations of dielectric constant $\delta\varepsilon$ have week dependence on the frequency, i.e., the main dependence on ω comes from the background dielectric constant ε. Therefore both the fundamental mode and second harmonic mode experience the same fluctuation $\delta\varepsilon$.

To find the intensity of the second harmonic mode we need to solve equations (14) and (15) with the nonlinear term. However, we know that to have the most efficient generation of the second harmonic mode the random resonator should satisfy the double resonant property. Therefore, we can reformulate the problem of finding the intensity of the second harmonic mode in a different way: we need to find the ring-shaped fluctuation of dielectric constant $\delta\varepsilon$ for which the probability determined by Eq. (4) is the largest under the condition that the corresponding resonator has a given quality factor and can trap the fundamental mode with angular momentum m and frequency ω and the second harmonic mode with angular momentum $2m$ and frequency 2ω. In this formulation we disregard the nonlinear properties of the media, therefore from this moment we consider Eq. (15) without the last term. Within this picture the intensity of the second harmonic mode is determined by the quality factor of the random resonator. In other words, we assume that the nonlinear term is small. This means that the last term in Eq. (15), which is of the order of $\chi\psi_1^2 \sim \chi(\mathrm{X}\mathcal{T}_1)^2$, is smaller than the value of the localized second harmonic mode, which is $\omega\mathcal{T}_2$. Therefore the nonlinear coefficient should satisfy the inequality

$$\chi \ll \mathcal{T}_2/\omega\mathcal{T}_1^2 \sim (\omega\mathcal{T}_1)^{-1}. \quad (16)$$

On substituting expressions $\psi_1 = (\rho)^{-1/2}\chi_1(\rho)$ and $\psi_2 = (\rho)^{-1/2}\chi_2(\rho)$ in Eqs. (14) and (15) and taking into account that $m \gg 1$ and $w \ll \rho_0$, we obtain the following one-dimensional equations:

$$\hat{\mathbf{L}}_1\chi_1 = \frac{d^2\chi_1}{dx^2} + \delta\varepsilon k_0^2 \chi_1 - \epsilon_1 k_0^2 \chi_1 = 0, \quad (17)$$

$$\hat{\mathbf{L}}_2\chi_2 = \frac{d^2\chi_2}{dx^2} + 4\delta\varepsilon k_0^2 \chi_2 - 4\epsilon_2 k_0^2 \chi_2 = 0, \quad (18)$$

where $x = \rho - \rho_0$ [see Fig. 1(a)]. The parameters ϵ_i, which can be considered as the "eigenvalues" of the corresponding modes, can be related to the Q factors of localized modes of random resonator.[8] Namely, $\epsilon_1 = \varepsilon_1(3\ln Q_1/2m)^{2/3}$, $\epsilon_2 = \varepsilon_2(3\ln Q_2/4m)^{2/3}$. These expressions follow from the quasiclassical equations for the tunneling rate through the trapping barrier. The shape of the trapping barrier is determined by the wave equation outside of the random resonator, i.e., at $x > w$. In this region the wave equations are of Airy type:

$$\frac{d^2\chi_1}{dx^2} = \epsilon_1 k_0^2 \left(1 - \frac{x}{d_1}\right)\chi_1, \quad (19)$$

$$\frac{d^2\chi_2}{dx^2} = 4\epsilon_2 k_0^2 \left(1 - \frac{x}{d_2}\right)\chi_2, \quad (20)$$

where the width of the decay region is given by $d_1 = \epsilon_1 k_0^2 \rho_0^3/2m^2$ and $d_2 = \epsilon_2 k_0^2 \rho_0^3/2m^2$.

To find the realization of fluctuation of dielectric constant $\delta\varepsilon$ we are using the optimal fluctuation approach.[19,20] Due to narrow spatial width of ring-shaped random resonator the correlator of random dielectric constant is effectively one dimensional with the kernel

$$K_0(x - x') = \int_{-\infty}^{\infty} dy K[\sqrt{(x - x')^2 + y^2}]. \quad (21)$$

Within the optimal fluctuation approach we search for the fluctuation $\delta\varepsilon$ for which the functional $|\ln W|$ has the smallest value, $|\ln W|_{\min} = S_m$, under the condition that Eqs. (17) and (18) are valid for some nonzero functions χ_1 and χ_2. This is a standard optimization problem with constrains. Taking into account constrains (17) and (18) we can write the correspond functional in the following form:

$$\Xi\{\chi_1, \chi_2, \delta\varepsilon\} = \ln W - \lambda_1 \int dx[\chi_1(x)\hat{\mathbf{L}}_1\chi_1(x)] - \lambda_2 \int dx[\chi_2(x)\hat{\mathbf{L}}_2\chi_2(x)]. \quad (22)$$

Performing optimization of functional (22) with respect to $\delta\varepsilon$ we obtain the expression for $\delta\varepsilon$ in terms of χ_1 and χ_2

$$\delta\varepsilon(x) = \int K_0(x - x_1)[\chi_1^2(x_1) + 4\chi_2^2(x_1)]dx_1, \quad (23)$$

where we introduced the following expressions for Lagrange multiplies, corresponding to constrains (17) and (18),

$$\lambda_1 = 2\pi\rho_0/(\Lambda k_0)^2,$$

$$\lambda_2 = 2\pi\rho_0/(2\Lambda k_0)^2.$$

In the present problem we can choose arbitrary values for Lagrange multipliers by rescaling the functions χ_1 and χ_2.[9]

Then we substitute Eq. (23) into Eqs. (17) and (18) and obtain the final system of equations

$$\frac{d^2\chi_1(z)}{dz^2} + \frac{\chi_1(z)}{(\epsilon_1 k_0)^2} \int K_0(z - z_1)[\chi_1^2(z_1) + 4\chi_2^2(z_1)]dz_1 = \chi_1(z), \quad (24)$$

$$\frac{d^2\chi_2(z)}{dz^2} + 4\frac{\chi_2(z)}{(\epsilon_1 k_0)^2} \int K_0(z - z_1)[\chi_1^2(z_1) + 4\chi_2^2(z_1)]dz_1 = 4\beta\chi_2(z), \quad (25)$$

where $\beta = \epsilon_2/\epsilon_1$ and we introduce dimensionless length $z = \sqrt{\epsilon_1} k_0 x$.

We solve the system of equations (24) and (25) using a variational method.[9] At first we notice that solution of Eqs.

(24) and (25) corresponds to a minimum of the following functional:

$$\Phi\{\chi_1,\chi_2\} = \int \left\{ \left(\frac{d\chi_1}{dz}\right)^2 + \left(\frac{d\chi_2}{dz}\right)^2 + \chi_1^2 + 4\beta\chi_2^2 \right\} dz - \int \frac{K_0(z_2 - z_1)}{2\epsilon_1} \prod_{j=1,2} \{[\chi_1^2(z_j) + 4\chi_2^2(z_j)]dz_j\}. \quad (26)$$

Then we introduce explicit expression for the correlation function of disorder. Namely, we assume that the correlator of the disorder has the Gaussian form

$$K(r) = \exp(-r^2/R_c^2), \quad (27)$$

Therefore the one-dimensional correlator (21) is

$$K_0(x) = \pi^{1/2} R_c \exp(-x^2/R_c^2). \quad (28)$$

Then with the given correlator K_0, we search for a solution of the variational problem corresponding to functional (26) in the following form:

$$\chi_1(z) = A_1 \exp[-\gamma_1 z^2], \quad \chi_2(z) = A_2 \exp[-\gamma_2 z^2], \quad (29)$$

where constants A_1, A_2, γ_1, and γ_2 should be considered as variational parameters. From Eq. (15) we can see that the generation of the second harmonic mode is determined by the overlap of the functions χ_1^2 and χ_2. To have the largest overlap we need to have the same spatial width of χ_1^2 and χ_2. This introduces an additional constrain in the variational problem. Namely, we have $\gamma_2=2\gamma_1$. Finally, in the problem we have only three variational parameters A_1, A_2, and γ_1. The next steps are straightforward. We substitute expressions (29) for χ_1 and χ_2 in the functional (26). Then we find numerically the minimum of the functional with respect to variational parameters A_1, A_2, and γ_1 for given values of disorder parameters and the Q factor of the resonator. With these optimal values of A_1, A_2, and γ_1 we find from Eq. (23) the optimal fluctuation of dielectric constant $\delta\varepsilon(x)$. Then we substitute the optimal $\delta\varepsilon(x)$ into Eq. (4) and find the probability of realization of random resonator $W_{\max}=\exp[-S_2]$. This probability determines the density of random resonators with the double resonant property within disordered dielectric film

$$\mathcal{N}_2 = \mathcal{N}_{2,0} e^{-S_2(kl,Q_1,\beta)}, \quad (30)$$

where $\mathcal{N}_{2,0}$ is the prefactor[10,21] and l is the transport mean free path of a photon, which is related to disorder parameters Λ and R_c. The argument of an exponent in Eq. (30) can be expressed as

$$S_2 = \xi\Phi_2(r_c,\beta), \quad \xi \equiv \frac{2^4\pi^{1/2}m}{3^{3/2}(\Lambda k_0 R_c)^2}\left(\frac{\epsilon_1^3}{\varepsilon_1}\right)^{1/2}, \quad (31)$$

where Φ_2 is the function of dimensionless correlation radius $r_c=\epsilon_1^{1/2}k_0R_c$ and parameter β.

A similar expression can be derived for the density of random resonators[8] which can trap only the fundamental mode

$$\mathcal{N}_1 = \mathcal{N}_{1,0} e^{-S_1(kl,Q_1)}, \quad (32)$$

where $\mathcal{N}_{1,0}$ is the corresponding prefactor. Here S_1 has the form $S_1=\xi\Phi_1(r_c)$. The function $\Phi_1(r_c)$ can be obtained from the above equations if we set χ_2 equal to zero and take into account only fundamental mode χ_1.[8]

The resonators which can trap both fundamental and the second harmonic modes constitute only part of the whole number of resonators which can trap the fundamental mode, i.e., there is a relation $\mathcal{N}_1 \geqslant \mathcal{N}_2$ or $S_2 \geqslant S_1$. Therefore, we can introduce the fraction of the resonators with the double resonant properties by the following expression:

$$\mathcal{F} = \mathcal{N}_2/\mathcal{N}_1 = e^{-(S_2-S_1)}. \quad (33)$$

This expression is valid with exponential accuracy, i.e., we assume that $\mathcal{N}_{2,0}/\mathcal{N}_{1,0} \sim 1$. In Eq. (33) the argument of exponent (S_2-S_1) has the form

$$(S_2 - S_1) = \xi[\Phi_2(r_c,\beta) - \Phi_1(r_c)] \equiv \xi\Delta\Phi(r_c,\beta). \quad (34)$$

Therefore the efficiency of generation of the second harmonic mode, which is proportional to the density of resonators with the double resonance properties, is determined by function $\Delta\Phi(r_c,\beta)$. When this function is zero, then all random resonators, which trap the fundamental mode, will contribute to efficient local generation of the second harmonic mode, i.e., they can trap the second harmonic mode also.

IV. RESULTS AND DISCUSSION

The results of the numerical calculation of function $\Delta\Phi(r_c,\beta)$ are shown in Fig. 2. The function depends only on two parameters. We can see from this figure that at fixed value of parameter β there are two different types of dependencies of $\Delta\Phi$ on correlation radius r_c. For $\beta<1$ and $\beta \gtrsim 1.4$ we have monotonic increase of $\Delta\Phi$ with r_c. For all values of correlation radius the function $\Delta\Phi$ is positive, i.e., it never becomes zero. For parameter β in the interval $1<\beta \lesssim 1.4$ the function $\Delta\Phi(r_c)$ is nonmonotonic and has zero minimal value at some correlation radius $r_c=r_{c,\min}(\beta)$. This point, i.e., the point at which $\Delta\Phi(r_c)$ becomes zero, is especially important since it means that all the resonators contribute to local generation of the second harmonic mode. As we can see from Fig. 2(b) the value $r_{c,\min}$ monotonically decreases with increasing parameter β within the interval $1<\beta<1.4$. The function $r_{c,\min}(\beta)$ covers all the values of correlation radius, from $r_{c,\min}=\infty$ at $\beta=1$ to $r_{c,\min}=0$ at $\beta \approx 1.4$.

At fixed value of correlation radius r_c, the dependence of $\Delta\Phi$ on parameter β is nonmonotonic, see Fig. 3. The function $\Delta\Phi$ has minimum at some value of β. The value of $\Delta\Phi$ at the minimum is always zero. Comparing with Fig. 2 we can tell that the position of minimum of $\Delta\Phi(\beta)$ is within the interval $1.4>\beta>1$.

The requirement of efficient generation of the second harmonic mode, i.e., $\Delta\Phi=0$, introduces the limitations on the possible values of the parameters of the random nonlinear media. To understand these limitations we rewrite parameter β in the following form:

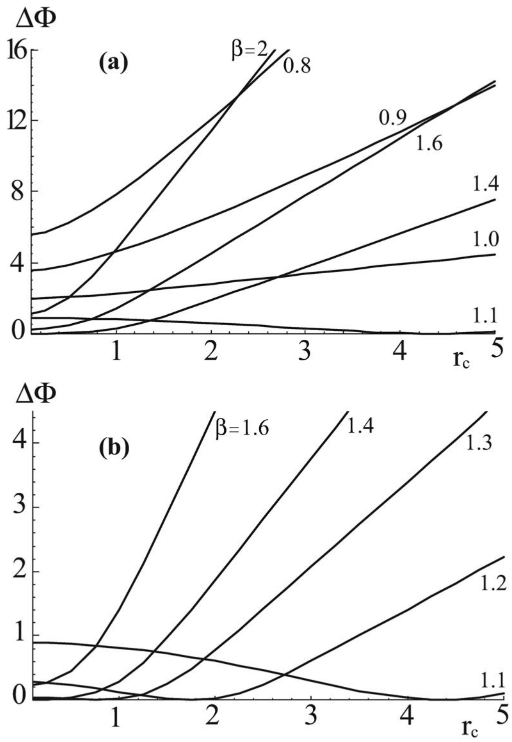

FIG. 2. A function $\Delta\Phi$, which characterizes the fraction of random resonator with the double resonant properties, is shown as a function of dimensionless correlation radius r_c (a) for different values of parameter β in the whole range of variations of β; (b) for parameters β in the interval, where $\Delta\Phi(r_c)$ has nonmonotonic dependence and becomes zero at some point.

$$\beta = \frac{\epsilon_2}{\epsilon_1} = 1 + \frac{\Delta\varepsilon}{\epsilon_1}, \tag{35}$$

where $\Delta\varepsilon = \varepsilon_1 - \varepsilon_2 = \varepsilon(\omega) - \varepsilon(2\omega)$ determines the phase mismatch between the fundamental and second harmonic modes in homogeneous media. Then relation (35) between β and $\Delta\varepsilon$ introduces the limitations on the possible values of $\Delta\varepsilon$. Since $\Delta\Phi$ can be zero only for $1<\beta<1.4$, we have $0<\Delta\varepsilon<0.4\epsilon_1$. For example, for resonator with parameters $\varepsilon=10$, $\ln Q=2$, $m=10$ we obtain $0<\Delta\varepsilon<1.8$.

The data shown in Figs. 2 and 3 are presented in the dimensionless units. Therefore, the parameter β and dimensionless correlation radius r_c depend on the parameters of random resonators, i.e., the Q factor, and on the parameters

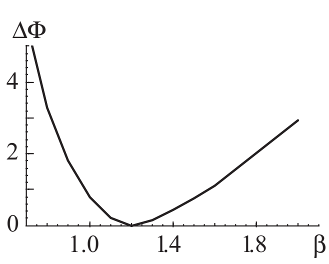

FIG. 3. A variable $\Delta\Phi$ is shown as a function of parameter β at $r_c=1.75$. The function $\Delta\Phi(\beta)$ has a minimum with zero value at $\beta=1.2$.

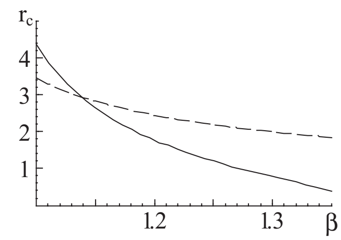

FIG. 4. Two dependencies are shown: one (solid line) is the relation (36) between r_c and β for different parameters of random resonators and for $k_0R_c=2$, and another (dashed line) is $r_c=r_{c,\min}(\beta)$, which determines the position of the point $\Delta\Phi(r_c,\beta)=0$. The intersection of two curves determines the optimal value of β and the optimal parameters of random resonator.

of random media. The parameters of random resonators enter this dependence through $\epsilon_1(Q_1)$ only. In the real systems the parameters of disorder are fixed and we change the quality factor of random resonators. Therefore, it is important to find the relation between β and r_c for a given random media, i.e., for a given correlation radius of disorder. This relation should not contain the parameters of random resonators, i.e., it should be valid for all resonators. To derive this relation we find ϵ_1 from expression $r_c=k_0R_c\epsilon_1^{1/2}$ and then substitute it into Eq. (35). Finally, we have the relation

$$r_c = k_0R_c\sqrt{\Delta\varepsilon/(\beta-1)}, \tag{36}$$

which is valid for a given random media and does not depend on the parameters of random resonators.

We can use relation (36) to find the parameters, i.e., the Q factor of the random resonator, which provide the most efficient frequency conversion, i.e., $\Delta\Phi=0$. Since we consider a given disordered system then r_c and β should satisfy Eq. (36). At the same time since we need to find the optimal resonator, i.e., $\Delta\Phi=0$, then we have the relation between r_c and β through the function $r_c=r_{c,\min}(\beta)$. Simultaneous solution of Eqs. (36) and $r_c=r_{c,\min}(\beta)$ determines the values of r_c and β and correspondingly the Q factor of random resonator. Although the function $r_c=r_{c,\min}(\beta)$ is universal, the expression (36) depends on the parameters of disorder through coefficient $k_0R_c\sqrt{\Delta\varepsilon}$. In Fig. 4 we show, as an example, this dependence at $k_0R_c=2$ together with function $r_c=r_{c,\min}(\beta)$. The intersection of two curves determines the optimal parameter β and correspondingly the parameters of optimal resonators.

The dependence $r_{c,\min}(\beta)$ is universal, and it behaves as $\simeq(\beta-1)^{-0.74}$ when β approaches 1. Since the β dependence in Eq. (36) is $(\beta-1)^{-0.5}$ there is always an intersection of these two dependencies. Therefore, for any disordered system there are random resonators, which provide the most efficient local conversion of the fundamental mode into the second harmonic mode. The parameters, i.e., Q factors, of such random resonators are fixed by the parameters of random media. This means that for any other Q factor the function $\Delta\Phi$ is greater than zero. Therefore only part of the whole number of random resonators, which can trap the fundamental mode, can trap the second harmonic mode at this quality factor.

Based on the above analysis we can estimate the density of the bright spots of the second harmonic mode. For example, we assume that the fundamental wavelength is $\lambda = 0.6\ \mu$m, the dielectric media has a weak disorder, i.e., $kl \approx 10$, and we are looking at the bright spots of the second harmonic mode with the local enhancement ≈ 500 compared to the average value. We also assume that the favorable conditions are achieved, i.e., $\Delta\Phi = 0$. Another crutial parameter of the system is the correlation radius of the disorder. The density of the bright spots has very strong dependence on the correlation radius of the disorder, i.e., kR_c. For $kR_c = 1$ the density of the bright spots is very small: $\mathcal{N}_2 \sim 10^{-19}\ \mu\text{m}^{-2}$. With increasing the correlation radius, i.e., with increasing the size of the scatterers, the density is highly increased. For example, for $kR_c = 2$ the density is $\mathcal{N}_2 \sim 10^{-2}\ \mu\text{m}^{-2}$.

In the derivation of the density of random resonators we assumed that the phase matching condition is satisfied, i.e., $m_2 = 2m$, and the overlap of the second harmonic mode and the fundamental mode in the radial direction is maximum, i.e., $\gamma_2 = 2\gamma_1$. Then we calculated the density of the resonators with a given trapping time. Strictly speaking, we need to calculate the density of the random resonators with a given local intensity of the second harmonic mode. The local intensity of the second harmonic mode, in addition to the factors $\mathcal{T}_2^2$ and $\mathcal{T}_1^4$ presented in Eq. (13), contains the factors, which determine the spatial overlap of the modes. Therefore the intensity of the second harmonic mode can be expressed in the following way:

$$I_2 \sim \chi^2 f_m f_\gamma \mathcal{T}_2^2 \mathcal{T}_1^4. \tag{37}$$

Here f_m characterizes the phase matching condition. This factor can also be considered as the factor which describes the spatial overlap of the second harmonic and the fundamental modes in the angular direction. The factor f_m is maximum when the phase matching conditions are satisfied, i.e., $m_2 = 2m$. The factor f_γ in Eq. (37) describes the spatial overlap of the modes in the radial direction. This factor is maximum when $\gamma_2 = 2\gamma_1$.

The reason that we assumed in our analysis that f_m and f_γ are maximum, is the following. The density of random resonators can be expressed as a product of two terms [Eq. (30)]: the exponentially small factor, which is determined by the exponent S_2, and the prefactor $\mathcal{N}_{2,0}$. In the present paper we calculate only the exponent, S_2, and do not consider the prefactor. This means that we consider the limit of exponentially small density of random resonators, i.e., $S_2 \gg 1$ and $S_2 \gg \ln \mathcal{N}_{2,0}$. In this limit the density of random resonators is completely determined by the exponent S_2, which itself has strong dependence on the trapping time $\mathcal{T}_1$. Then to have the largest density of random resonators we need to have the smallest value of S_2 and correspondingly the smallest value of the trapping time. The smallest trapping time should be defined under the condition that the local intensity I_2 of the second harmonic mode has a given value. Then we can see from Eq. (37) that the trapping time is the smallest when the factors f_m and f_γ have the largest values. Therefore, in our analysis we assumed the perfectly matching conditions, i.e., $m_2 = 2m$ and $\gamma_2 = 2\gamma_1$, since these conditions provide the smallest possible value of the trapping time under a given value of the local intensity of the second harmonic mode.

The deviations of the factors f_m and f_γ from their maximum values, i.e., violation of the conditions $m_2 = 2m$ and $\gamma_2 = 2\gamma_1$, make the contributions to the prefactor in Eq. (30). In the above analysis we did not take into account the frequency dependence of the fluctuating part of dielectric constant, so both the fundamental and second harmonic modes experience the same effective potential due to fluctuations of dielectric constant. In the real system the fluctuating part of dielectric constant depends on the frequency, but there is strong correlation between the values of dielectric constant at fundamental and second harmonic frequencies, e.g., if dielectric media is a system of random dielectric particles. The frequency dependence of fluctuating part of dielectric constant modifies qualitatively the function $\Delta\Phi$, but the main qualitative results will remain the same. Namely, for each random media there are some parameters of random resonators, i.e., quality factor of the resonators, for which the number of resonators, which can trap the fundamental mode, is equal to the number of resonators, which can trap both the fundamental mode and the second harmonic mode.

The random resonators with the double resonant property produce strong local enhancement of the second harmonic mode. Such local enhancement can be observed in the spatial distribution, i.e., speckle pattern, of the second harmonic field at the surface of the disordered film. The positions of the bright spots in the speckle pattern at the second harmonic frequency determine the position of random resonators. Comparison of the density of the bright spots at the fundamental frequency and at the second harmonic frequency can determine the fraction of random resonators which can trap the light at the fundamental and the second harmonic frequencies.

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