Generation of Light in Media with a Random Distribution of Nonlinear Domains

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We show that the second order nonlinear generation of light, a process that it is assumed to require highly ordered materials, is also possible in structures of randomly oriented nonlinear domains. We explain theoretically why in such disordered structures the efficiency of the nonlinear generation of light grows linearly with the number of domains. Moreover, a higher degree of disorder, obtained when the dispersion is made very large, has no negative effect for the nonlinear light generation. In such conditions, light generation is shown to be equally efficient for any average size of the domains and also to grow linearly with respect to the number of domains.

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It is commonly accepted that quadratic nonlinear processes, such as parametric generation or amplification, require the use of materials with a high degree of ordering. In some occasions, such ordering is found at a nanoscale or molecular scale, and in other cases, the order is at a micron scale. When such ordering is not intrinsic to the material, one may introduce a periodical distribution within the nonlinear material to, for instance, compensate the phase mismatch. In that event, the final material would be, in general, composed of two types of domains, distributed periodically across the entire material, one with a given nonlinear coefficient, and the other with the same coefficient with opposite sign [1]. The length of these domains must be one coherence length (ℓ_c), a length that for a large number of nonlinear materials ranges from several hundreds of nanometers to several tens of microns. Then, the nonlinear interaction becomes what is known as quasiphase matched, which leads to a quadratic growth of the second harmonic intensity in terms of the number of domains. In principle, one would expect that small deviations from the adequate period, or some dispersion in the size of the domains, would lead to a cancellation of the coherent nonlinear process of three wave mixing. However, very recently it was observed that with polycrystalline samples fabricated with a random orientation of zinc selenide (ZnSe) crystalline domains, when the average size of the domains was close to $1\ell_c$, difference frequency generation grew linearly with the total length of the sample [2]. Similar observations were reported some years ago from $Sr_{0.6}Ba_{0.4}Nd_2O_6$ needlelike crystalline domains [3] and with the use of rotationally twinned crystals of ZnSe [4,5]. In all these observations, the efficiency of the process seemed to be strongly linked to an average size of the domain close to the optimal value for quasiphase matching with periodical inverted domains.

In the present work, we study the process of phase matching to compensate the material dispersion in the refractive index in materials where there is no structural ordering. Here, we consider one-dimensional (1D) structures composed of planar layer domains with a well-defined orientation of the nonlinear susceptibility within the domain. Such domains, however, are randomly ordered and their thickness may vary with a Gaussian distribution around a given average size. In other words, the entire structure exhibits no ordering with respect to the orientation of the dipoles and the domains are allowed to have any possible thickness. Contrary to what one might expect, we observe that an increase in the amount of disorder is not necessarily detrimental with respect to the efficiency of a second harmonic generation (SHG) process. Moreover, the linear growth of the second harmonic (SH) intensity with respect to the number of domains is seen when the average size of the domains is close to $1\ell_c$, but also for any other average thickness of such domains. As we shall see below, we are able to establish a clear link between the disorder inherent to the structure and the linear growth of the intensity with respect to the number of domains.

The 1D structure we considered is made of stacks of planar layers of a nonlinear homogeneous material, infinite in the xy plane, and with a finite thickness in the z direction. The refractive index and dispersion was assumed to be the same for all layers. In each layer the nonlinear susceptibility is determined by the nonlinear polarizability of the dipole, the density of dipoles, and the orientation of such dipoles with respect to the orientation defined by the 1D planar structure [6]. Within a given layer all such dipoles pointed in a certain direction; however, there was no correlation between the dipole orientation within that given layer and that same orientation within any other layer. Moreover, we assumed that the layer thicknesses follow a Gaussian distribution around a certain average thickness and with the corresponding standard deviation. Once again, there was no correlation between the thicknesses of a given layer with respect to any other layer. In such a structure, we considered SHG from an incident plane wave with the wave vector parallel to the z axis while the electric field was linearly polarized on the xy plane. The SH electric field was assumed to be polarized in the same direction as the nonlinear polarization took when projected on the xy plane [7]. Under the assumption of harmonic waves, one may determine SHG from the entire stack by solving the wave equation of the SH electric field amplitude within each one of the layers. In our case, where the refractive index of all the stacks is the same, the nonlinear polarization source may be written as $P^{nl}_{2\omega,n} = \varepsilon_0 \chi^{(2)}_n E^2_\omega$ at the nth layer, where $\chi^{(2)}_n$ is the second nonlinear susceptibility of the nth layer and E_ω is the fundamental electric field.

Under the nondepleted pump approximation one may obtain an analytical expression for the slowly varying part of the SH electric field amplitude within the nth layer at z' generated by the fundamental field:

$$E_{2\omega,n}(z') = -\frac{2\omega^2\chi^{(2)}_n}{k_{2\omega}\Delta k}E^2_\omega[e^{i\Delta k z'} - 1], \qquad (1)$$

where z' ranges from 0 to d_n (the thickness of the nth layer), and $\Delta k = k_{2\omega} - 2k_\omega$ is the phase mismatch being $k_{2\omega}$ and k_ω the z component of the SH and fundamental field wave vectors within the nonlinear layer, respectively. The total field at each layer, the product of Eq. (1) with the rapidly varying phase, is used together with the transfer matrix formalism applied to both frequencies to determine the generated field at 2ω across the entire structure.

In a numerical simulation, the disordered 1D structure was assumed to be made of a material with the same index of refraction and dispersion as in ZnSe when the incident field was tuned at 1064 nm and SH at 532 nm. In the numerical calculations, unless otherwise specified, we assumed that $\chi^{(2)}E_\omega = \sqrt{2} \times 10^{-4}$. A random orientation of the nonlinear susceptibility in each layer implies that the normalized component of the nonlinear polarization in the direction of the incident electric field ranges from 1 to -1. To implement such configuration numerically, we used a random number generation algorithm, which provides, with equal probability, a number between 0 and 1 [8]. Finally, the nonlinear susceptibility $\chi^{(2)}$ of the material was multiplied by the cosine of the output of the random generator multiplied by π. As mentioned earlier, in addition to a random orientation of the $\chi^{(2)}$ within each layer, the thickness of the layers of any given 1D structure exhibited a random Gaussian distribution. Numerically, to generate a random deviation of the thickness with a normal distribution we used the Box-Muller method [9,10]. Eventually, to evaluate the performance of a given type of structure, we performed an average over 200 structures of the same kind. One may view this configuration as a set of 200 parallel tubes, where each tube is made of slices of different nonlinearity and thicknesses. Note that, experimentally, such an average would be performed automatically, since in most cases the transverse dimensions of the incident beam are such that, as in Refs. [2,3], several tens of domains are illuminated simultaneously.

If we consider SHG in a given disordered structure as a function of the number of layers, we observe, as shown in Fig. 1, that the efficiency cannot be correlated to the number of layers. However, when we perform the average over 200 structures, the SHG efficiency grows linearly with the number of layers when the average layer thickness equals ℓ_c and the percentage coefficient of variation (CV) in the thickness of each layer of the structures is 1% [11], as seen also in Fig. 1. In a random orientation of the domains there are four possible sequences of bilayers: up-up, up-down, down-up, and down-down. The second and third will always give a positive contribution, while the first and fourth will give a very small contribution: zero if the nonlinear coefficients of the adjacent layers are exactly the same. A given sequence of a large number of layers will combine these four possible bilayers in a way that the efficiency of the entire structure will range from zero to the efficiency found for perfect quasiphase matching. Then, when we average over a large number of stacks there is a noncoherent addition of the contribution to SHG from all bilayers, which leads to the linear rather than quadratic growth of the efficiency. This linear growth is in agreement with the experimental observation of the linear growth reported in Ref. [2]. If the CV is increased up to 32%, as shown in Fig. 2, the linear growth of the efficiency is maintained. Such efficiency is shown also in Fig. 2 when the average size of the domains is $5\ell_c$ and $10\ell_c$. Although, the rate of growth in both such cases is reduced with respect to the structure with domains around $1\ell_c$, there is no significant difference between them. In other words, when the average size of the domain increases and the dispersion is large, the efficiency in terms of the number of domains becomes independent of the average domain size. We used the parameters of ZnSe [12] with CV = 1% to estimate that with less than 30 cm of this material, and 2 GW/cm^2 of input peak intensity, one would reach 10% conversion to SH light.

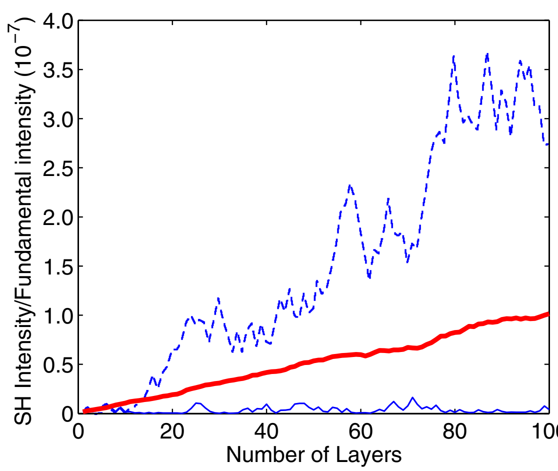

FIG. 1 (color online). SH intensity as a function of the number of layers normalized to the intensity of the incident field when the coefficient of variation (CV) is 1% and the average layer thickness equals ℓ_c. The dashed line and the solid thin line correspond to two different random structures, while the solid thick line corresponds to an average over 200 different random structures.

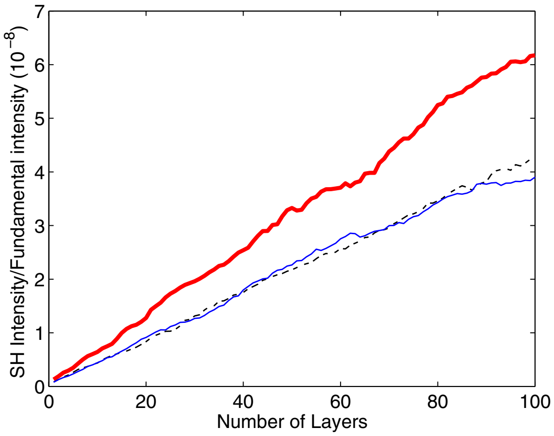

FIG. 2 (color online). SH intensity averaged over 200 different random structures normalized to the intensity of the incident field as a function of the number of layers when the CV is 32% and the average layer thickness equals ℓ_c (solid thick line), $5\ell_c$ (solid thin line), and $10\ell_c$ (dashed thin line).

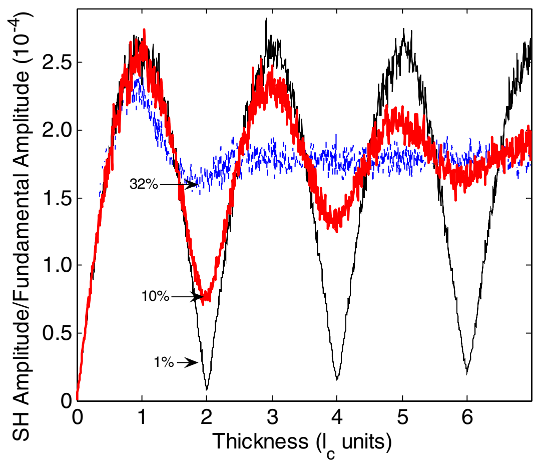

FIG. 3 (color online). SH field amplitude normalized to the amplitude of the incident field as a function of the average thickness of the domains when the CV is 1% (solid thin line), 10% (solid thick line), and 32% (dashed line).

Although the linear behavior is found for small and large layer thickness dispersion, it is not apparent why the efficiency, at least when the dispersion is large, seems to be independent of the average thickness of the domain. To resolve this issue we studied the efficiency of a stack of 100 layers as a function of the average thickness of the domain. First, we considered structures with a random orientation of the dipole associated with the $\chi^{(2)}$ but a very small variation of the thickness of the layers, CV $= 1\%$. As above, the SH efficiency was determined after averaging over 200 structures. As shown in Fig. 3, we obtained a maximum generation when the thickness of the domains is close to 1 or any other odd multiple of ℓ_c. As expected, when the thickness of the domain is an even number of ℓ_c, independent of the sequence of layers one may get, SH is completely destroyed within that one layer, which results in sharp zeros of generation also seen in Fig. 3. However, if the dispersion in the thickness of the layers is increased up to CV $= 10\%$, we observed that, simultaneous to a reduction of the amplitude of the field generated for thickness with mean values near or equal to an odd multiple of ℓ_c, the nonlinear conversion is also possible for thicknesses near to an even multiple of ℓ_c. The relative maxima decrease while the relative minima increase as the multiple becomes larger. These two effects combined lead to a constant value for the SH amplitude as the thickness of the domains increases. Such constant amplitude, which is approximately 0.7 times the maximum amplitude, is seen for almost any average length of the domain when the dispersion is very large. For instance, when CV $= 32\%$ such constant conversion already appears when the domain thickness is less than 3 times ℓ_c (cf. Fig. 3). Such quenching of the SHG oscillations as a function of the average size of the domain may be attributed almost entirely to what happens within one nonlinear layer independent of the rest. When the thickness is close to an odd multiple of the ℓ_c, the extra or missing portion of material results in a diminished generation. On the contrary, when the mean value of the thickness is close to an even multiple of the ℓ_c, both the extra and the missing amount of thickness result in a more efficient generation since the thickness of the layer becomes closer to an odd multiple of ℓ_c. In the case of extra thickness, this amount results in the generation of a net quantity of light. On the other hand, with shorter domains not all the light generated in the previous one is canceled in the following one. Note that, as seen in the Fig. 3, the SHG remains essentially the same as when the average domain thickness is equal to $(n - 1/2)\ell_c$, where n is any positive integer. In other words, for structures with a non-negligible layer thickness dispersion, SHG oscillates as a function of the layer thickness but approaches the conversion one obtains when the thickness is equal to half $1\ell_c$, whatever the dispersion is. When the average thickness of the domains is less than $\ell_c/2$, the CV has negligible influence. In that case, almost no domain, even when the dispersion is as high as 32%, would be longer than $1\ell_c$; therefore all of them give a net contribution to the generation of light.

We also considered a structure that combines two types of materials with a different average layer thickness, one with the dipoles pointing up with respect to the direction of the field polarization and the other pointing down. In this case, we assumed there were no intermediate values for the dipole orientation. When we considered the efficiency with respect to the relative layer thickness between these two types of layers, we observed that a maximum SHG appears when the average thickness of both types of layers is centered at $1\ell_c$, as seen in Fig. 4(a) where the efficiency is shown as a function of both layer thicknesses. A closer inspection of that figure indicates that the maximum generation is achieved within a diamond shaped area around this central point. One of the diagonals of this diamond corresponds to layer pairs where the thicknesses of both

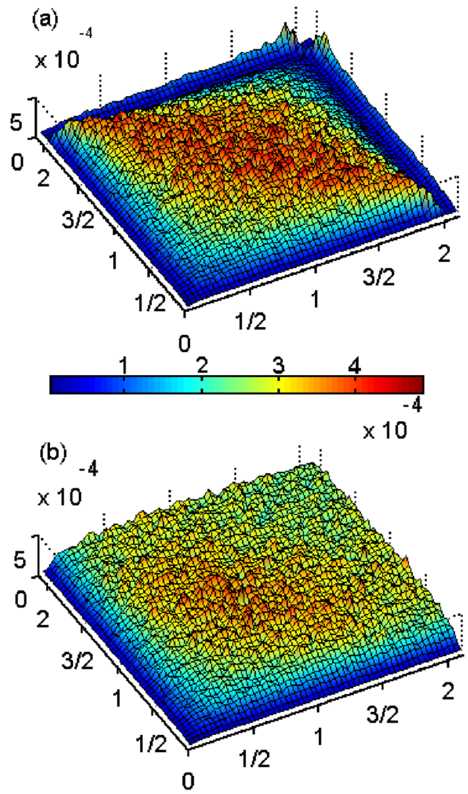

FIG. 4 (color online). SH field amplitude normalized to the amplitude of the incident field (z axis) as a function of the average thickness of the layers in units of ℓ_c with the nonlinear polarization pointing down (x axis) and of the layers with nonlinear polarization pointing up (y axis): (a) when the CV is 1% and (b) when the CV is 32%. Average here is over 50 structures.

layers combined is equal to 2 times ℓ_c, while the other diagonal corresponds to layers of equal thickness as one would expect from the results shown in Fig. 3. One may conclude that for a more efficient generation in an actual structure fabricated combining two types of domains the average size of these two different types does not necessarily have to be equal. This positive effect is more apparent when the dispersion in the size of the domains is increased. As shown in Fig. 4(b), SHG as a function of the size of both domains turns into a large plateau with an average value of the SH amplitude equal to the one found at $\ell_c/2$ for the structure with no dispersion. In summary, when disorder is large, light generation is found for almost any average size of the domains and for any relative size between the domains that point up or down.

To conclude, we have demonstrated that highly coherent processes such as SHG are possible in materials with a high degree of disorder. When the average size of the domains is larger than $1\ell_c$, and there is no preferred orientation for the domains, increasing the disorder by increasing the dispersion on the domain size is not detrimental for SHG. On the contrary, when the average size of the domains increases, SHG becomes independent of the size of such domains and also grows linearly with respect to the number of domains. Moreover, our theoretical prediction of linear growth is in agreement with the experiments performed in the past with random microcrystalline structures. Using parameters typical from some nonlinear materials, we show that an efficient conversion would be possible using nonlinear structures several cm long.

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[6] See, for instance, Joseph Zyss, in *Molecular Nonlinear Optics*, edited by Joseph Zyss (Academic Press, San Diego, 1994), Chap. 3, p. 105.
[7] In order to avoid any nonrelevant analytical or numerical complexity we assume that the nonlinear interaction is dominated by only one element of the nonlinear polarizability tensor which does not change the polarization of the SH with respect to the fundamental wave.
[8] Marsaglia's generator, the random number generator algorithm we used, may generate a total of 2^{1430} uncorrelated numbers within one loop of a numerical code. In our case the subroutine is called a maximum of 12 times, approximately 2^{1426} times orders of magnitude below the threshold that would repeat itself. This subroutine is often called pseudorandom; see, for instance, *Numerical Recipes in FORTRAN: The Art of Scientific Computing* (Cambridge University Press, Cambridge, 1992).
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