Second-harmonic generation in local modes of a truncated periodic structure

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We present an experimental study of the generation of second-harmonic light in a one-dimensional periodic structure truncated by the introduction of a defect in the central period. We observed an enhancement of the nonlinear interaction in the vicinity of the defect when the second-harmonic wave was excited for modes within the forbidden zone or stop band. We also observed an enhancement near the band edge, where the group velocity approaches zero. Second-harmonic generation is completely suppressed for local modes within the forbidden band other than the defect mode.

Selection of electromagnetic modes in a periodic dielectric material may be used to obtain sharp optical features such as suppression and enhancement of the radiation of oscillating dipoles. Suppression of electromagnetic modes resonant with the radiative frequency of a molecular dipole led to the observation of partial inhibition of spontaneous emission.[1] This selection may be accomplished in periodic distributions of dielectric materials that exhibit a strong reflection for a range of wavelengths centered at any wavelength that satisfies the Bragg condition. In analogy with electronic band gaps of solid-state theory, in recent years three-dimensional dielectric periodic structures have been called photonic band-gap (PBG) structures.[2] The language of PBG theory may be particularly useful when defects that break the perfect periodicity of the lattice are included.[3] The removal of some dielectric material from a PBG structure produces a highly localized defect mode analogous to an impurity state.[3] Recent experimental measurements have determined a high energy-density distribution near a defect of a two-dimensional periodic structure in the microwave domain of the spectrum.[4] This high local intensity may also strongly alter the nonlinear interaction with the material. Recent theoretical calculations demonstrated intensity-dependent pulse transmission and reflection in one-dimensional periodic structures.[5]

In this Letter we present an experimental study of the environmental effect on the second-harmonic (SH) radiation of a slab of nonlinear material embedded in a truncated multilayer stack of dielectric material. The localization of energy near a defect mode, as well as the reduction in the group velocity near the band edge,[6] will result in sharp resonances for the nonlinear interaction in the vicinity of the defect.

In our experiments we used a truncated periodic structure, depicted in Fig. 1, formed by two identical multilayer stacks separated by a small air gap that acts as the defect (or impurity). Each stack consists of 27 alternating amorphous dielectric layers of high index $n_1 = 1.95$ and low index $n_2 = 1.46$ and of thicknesses 75.5 and 101 nm, respectively, deposited upon a glass substrate. Such a structure strongly reflects light at normal incidence in the wavelength range from 540 to 600 nm. A range of total reflection, known as the Bragg stop band or forbidden band, is also seen when the wavelength is maintained constant at some value close to the wavelength that satisfies the Bragg condition and the angle of incidence is varied. The length of the defect or air gap separating both stacks was controlled by a micrometer stage that moved one of the multilayer stacks relative to the other. A monolayer of oriented Malachite Green (MG) chromophores was adsorbed on the surface facing the air gap of one of the multilayer stacks. The stack was immersed in a 5.0×10^{-4} M solution of MG in 1-propanol and removed from the solution at a constant speed of 5 mm/min.[7] Second-harmonic generation (SHG) in the dipole approximation is possible because of the lack of inversion symmetry at the surface layer of MG.[8]

The SH intensity reflected from the surface monolayer embedded in the one-dimensional periodic structure described was measured after excitation with 35-ps pulses from an active–passive mode-locked Nd:YAG laser. The average energy of the pulse was 5 mJ, and the polarization of the incident beam was set to be either parallel (TM) or perpendicular (TE) to the plane of incidence. Symmetry considerations indicate that the SH reflected beam from the structure is TM polarized in both cases.[9] A reflection geometry was used to avoid interference with the SH field generated in the bulk of the substrate. The incident beam was focused to a beam diameter of 2 mm onto the truncated periodic structure, which was mounted upon a rotating stage. The reflected SH signal from the structure was measured with an R212 Hamamatsu photomultiplier preceded by heat absorbing and interference filters.

For the periodic structure described, a gap is found at the SH frequency between 15 and 63 deg when the field is TM polarized. Peaks of high transmission

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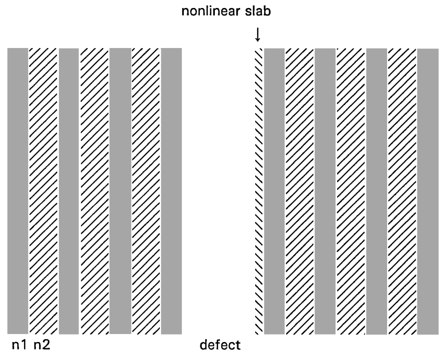

Fig. 1. Two one-dimensional stacks of alternating layers of indices n_1 and n_2 separated by an air gap or defect layer. The monolayer of nonlinear molecules is pictured on the front surface of the multilayer stack at the right. The widths of the dielectric layers and the defect layer are drawn to scale of the experimental parameters.

appear within the stop band when an air gap or defect is introduced into the structure. The field at the fundamental frequency is nonresonant with the wave vector of the periodic structure and thus is highly transmitted and only slowly modulated as a function of the angle of incidence.

When the light pulse at the fundamental frequency is transmitted through the truncated periodic structure, light at the SH will be generated at the monolayer of MG molecules. The mirrorlike periodic distribution of dielectric material in both sides of this monolayer will strongly alter the nonlinear interaction. Experimental measurements of the SH intensity reflected from the surface monolayer of nonlinear molecules through the multilayer stacks as a function of the angle of incidence are shown in Fig. 2, with the incident fundamental field TM polarized. A sharp resonance at 29 deg corresponding to the excitation of the SH field in a local mode within the forbidden band may be seen in Fig. 2, as well as another resonance at the large angle edge of the stop band at 63 deg. We performed additional experiments to determine the angular dependence of SHG when the multilayer stacks are separated by a distance much larger than the coherence length of the laser pulse and all coherent effects disappear. The reflected SH intensity in such conditions is shown in Fig. 3 as a function of the angle of incidence.

A comparison of Figs. 2 and 3 indicates that at an angle of 29 deg the generated SH intensity is six times larger when the nonlinear interaction occurs in a mode of the microresonator formed by the two multilayer stacks. The air gap separating the two stacks results in the appearance of a defect mode within the forbidden band or impurity level. The high energy density of this local mode is responsible for the enhancement of several times the nonlinear interaction in the vicinity of the defect. In the language of PBG theory, this sharp resonance corresponds to the excitation of the SH oscillation in a donor mode.[3] In a one-dimensional lattice the position of this bound state in the angular spectrum shown in Fig. 2 is a function of the size of the defect.

Near the band edge the bending of the electromagnetic wave dispersion curve slightly above or below the forbidden zone indicates that the group velocity approaches zero, giving rise to an increased effective path length and a Van Hove–type singularity in the photon density of states for a one-dimensional lattice. In our experiment this leads to an enhancement of the SH radiation at the angle of 63 deg, as shown in Fig. 2. This enhancement of the nonlinear interaction, predicted by Bloembergen and Sievers,[10] is a result of the periodicity built into the material, and its location in the angular spectrum shown in Fig. 2 is essentially independent of the size and position of the defect.

The nonlinear interaction in modes lying within the forbidden zone other than the defect mode is inhibited by a destructive interference among the forward-generated wave, the backward-reflected wave,

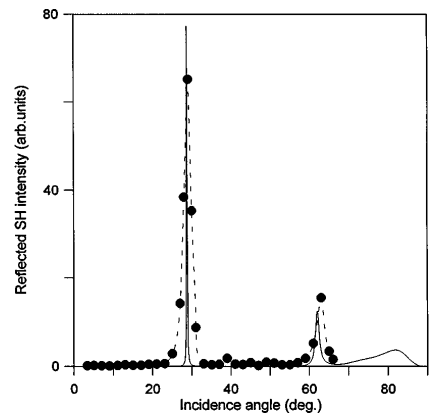

Fig. 2. Reflected SH intensity as a function of the angle of incidence, from the multilayer stack described in the text. The angle is given relative to the normal of the multilayer stacks. The filled circles indicate the experimental data; the dashed curve is a guide for the eye. The continuous curve corresponds to the numerical prediction of the theoretical analysis. The scale for the theoretical curve is reduced by a factor of 10 relative to the scale of the theoretical curve in Fig. 3.

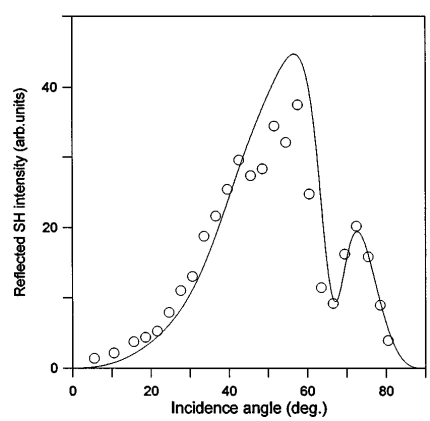

Fig. 3. Reflected SH intensity when the separation among the multilayer stacks is larger than the coherence length of the laser pulse. The open circles indicate the experimental data, and the continuous curve corresponds to the numerical prediction of the theoretical analysis. The scales for the experimental data in this figure and Fig. 2 are the same.

and the dipoles oscillating at the SH frequency. As one can see by comparing Figs. 2 and 3, SHG is completely suppressed for modes other than the defect mode within the forbidden band. This suppression of the oscillation at the SH frequency corresponds to the inhibition of the radiation from a classical dipole source.[11]

The appearance of a resonance structure may be described by a model including the interference effects that arise from multiple reflections in the mirrorlike structure. In a theoretical analysis of the experimental results we considered first the generation of SH light in a nonlinear slab of thickness δ approaching zero by solving the linear wave equation with a nonlinear source term and setting the appropriate boundary conditions at the interfaces.[12] The electromagnatic field at the SH frequency, 2ω, inside the nonlinear slab is given by the sum of an incident wave and a reflected wave, both solutions of the homogeneous wave equation, and a source wave resulting from the nonlinear interaction with the wave from the fundamental frequency. We have neglected the source wave resulting from the nonlinear interaction with the fundamental reflected wave. The index of refraction of the nonlinear slab is taken to be the index of the dielectric layer where it is absorbed. In the limit $\delta < 2\pi c/\omega$ we may obtain a simple expression for the source field in terms of the nonzero components of the nonlinear polarization $\mathbf{P}^{\mathrm{NL}}$, which are determined after contraction of the nonlinear susceptibility $\chi^{(2)}$ and the tensor E^2, a solution of the homogeneous wave equation for the wave at the fundamental frequency. Surface molecular layers deposited by the dipping technique described above may be considered rotationally symmetric about an axis normal to the plane of the layer. By symmetry considerations we may see that there are only three nonvanishing tensor elements, $X_{ZZZ}^{(2)}$, $X_{ZXX}^{(2)}$, and $X_{XZX}^{(2)} = \chi_{YZY}^{(2)}$.[9] The generated field propagating in the forward direction will be partially reflected at each interface of the periodically layered structure to form the reflected SH wave. The field in each layer of the stack is written as the sum of an incident and a reflected wave. By setting the input field at 2ω on both sides of the truncated periodic structure to zero we may solve for the total transmitted and reflected SH waves by the transfer matrix method.[13]

A numerical solution of the reflected SH wave is shown in Fig. 2, when the air gap separating the two multilayer stacks in the direction of propagation was $d/\cos\,\theta = 286$ nm. The length of the gap is d, and the angle of incidence is θ. The relative position and the relative intensity of both resonances are in close agreement with the experimental measurements. The numerical prediction of an enhancement of 90 times, determined by a comparison of the theoretical curves from Figs. 2 and 3, is considerably larger than the enhancement observed experimentally. This discrepancy probably arises from the spreading of the laser beam, diffusion by imperfections in the multilayer stacks, and imperfect parallelism of the two stacks.

The region of nonlinear interaction in surface SHG is limited to a few molecular or atomic layers. Given this confinement of the interaction, surface SHG in the vicinity of a defect could be adopted as a powerful tool for determination of the intensity space dependence in localized modes for electromagnetic waves in the visible region of the spectrum. On the other hand, enhancement of SHG in defect modes of the mirror structure could be particularly useful in the implementation of frequency doublers in vertical cavity surface-emitting lasers. When considering a nonlinear material distributed along the entire multilayer stack, one could expect an exponential growth of the SH amplitude as a consequence of the excitation of the SH wave in a local mode in this microresonator and of phase matching provided by the bending of the electromagnetic wave dispersion curve.[10,14]

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