Second-harmonic generation in hydrogenated amorphous-$Si_{1-x}N_x$ doubly resonant microcavities with periodic dielectric mirrors

S. Lettieri,[a] F. Gesuele, and P. Maddalena
C.N.R.–INFM - COHERENTIA, Dipartimento di Scienze Fisiche, Università di Napoli "Federico II", Via Cintia, I-80126 Napoli, Italy

M. Liscidini and L. C. Andreani
Dipartimento di Fisica "A. Volta," Università di Pavia, via Bassi 6, I-27100 Pavia, Italy

C. Ricciardi, V. Ballarini, and F. Giorgis
Dipartimento di Fisica, Politecnico di Torino, C.so Duca degli Abruzzi 24, I -10129 Torino, Italy and Laboratorio Materiali e Microsistemi (ChiLab), Lungo Piazza d'Armi 6, I-10034 Chivasso (Torino), Italy

(Received 12 May 2005; accepted 10 September 2005; published online 4 November 2005)

We report on the realization of microcavities made of hydrogenated amorphous-$Si_{1-x}N_x$ multilayers, with dual-wavelength periodic dielectric mirrors in order to obtain first and second order stop bands. Optical second-harmonic generation with simultaneous resonance for the pump and harmonic waves is demonstrated at finite values of the angle of incidence. The results are in good agreement with a theoretical calculation of the harmonic generation process based on a nonlinear polarization localized at the interfaces between different amorphous layers. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2125112]

Optical second-harmonic generation (SHG) in planar microcavities is a subject of growing interest in recent years. The long-term goal is to obtain large nonlinear conversion efficiency in a compact device. Simultaneous resonance for the pump and harmonic waves has been achieved with external cavity configuration[1,2] or in a monolithic cavity with nonperiodic mirrors.[3] Dual-wavelength periodic dielectric mirrors have been introduced in Refs. 4 and 5 and doubly resonant SHG in microcavities with such mirrors has been theoretically studied.[4,6,7]

Distributed Bragg reflectors (DBRs) and microcavities based on hydrogenated amorphous silicon nitride (a-$Si_{1-x}N_x$:H) can be produced by modulating the refractive index, that increases as a function of the nitrogen content.[8,9] SHG in silicon nitride microcavities with the pump wave being resonant with the Fabry-Pérot mode has been recently demonstrated with valuable enhancing of the optical nonlinearity.[10]

In this work we show the results of SHG analysis on a-$Si_{1-x}N_x$:H microcavities which were designed to be simultaneously resonant for pump and harmonic waves. We show that an enhancement of the nonlinear conversion occurs due to the double resonance, which can be maximized by tuning the angle of incidence. The harmonic generation process is theoretically modeled by a nonlinear transfer matrix method[11] suitably generalized for describing the surface $\chi^{(2)}$ processes. The present work represents, to the best of the author's knowledge, the first realization of doubly resonant SHG in microcavities with dual-wavelength periodic DBRs. Furthermore, we show that SH enhancement occurs even in systems where the nonlinear susceptibility is dominated by a surface (rather than a bulk) contribution, as is shown explicitly for the present a-$Si_{1-x}N_x$:H multilayers.

All-amorphous silicon nitride microcavities were grown by plasma enhanced chemical vapor deposition (PECVD) on 7059 Corning glass substrates. The composition of the a-$Si_{1-x}N_x$:H layers was controlled by operating on the ammonia fraction present in a SiH_4+NH_3 plasma. Their thickness was estimated taking into account the growth rate calculated by homogeneous thin films previously grown, while the composition ($x=N/Si+N$) of the alloys has been estimated by considering their refractive indices n, through a calibration curve $n(x)$ obtained by Rutherford backscattering spectrometry and optical interferometry performed on some selected a-$Si_{1-x}N_x$:H specimens.

A layout of the microcavity is shown in Fig. 1, upper panel. The structure parameters are as follows: $L_1=73$ nm(a-$Si_{0.57}N_{0.43}$:H), $L_2=219$ nm(a-Si_3N_4), $L_c=613$ nm(a-$Si_{0.45}N_{0.55}$:H). The design has been done by using the frequency dependent complex refractive indices determined by spectroscopic ellipsometry and interferometry.[12]

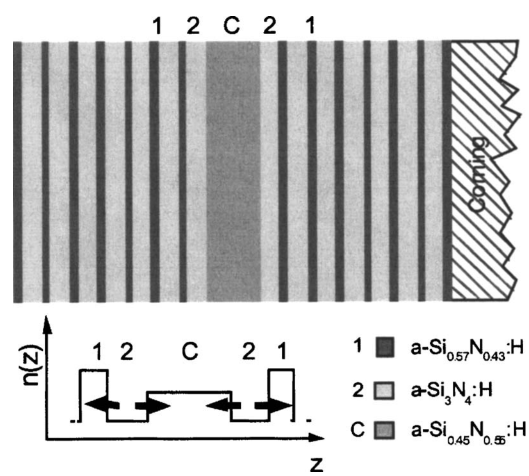

FIG. 1. Upper panel: layout of the microcavity structure, made of a defect layer "c" embedded between two dielectric mirrors made of layers "1" and "2." Lower panel: schematic profile of the refractive index. The arrows at the interfaces denote the direction from low to high refractive index, determining the sign of the $\chi^{(2)}$ components.

[a]Author to whom correspondence should be addressed; electronic mail: lettieri@na.infn.it

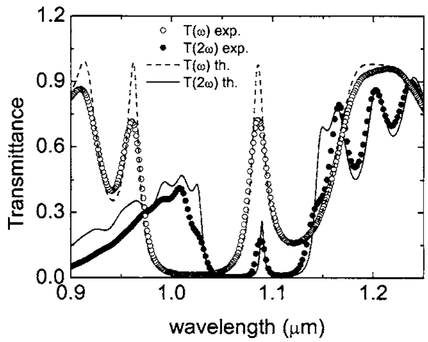

FIG. 2. Linear transmittance for *p*-polarized light at an incidence angle of 40°, in the spectral region of the pump wave (open dots and dashed line) and of the second-harmonic wave (full dots and solid line), as a function of the pump wavelength. Points: experimental results. Lines: theory.

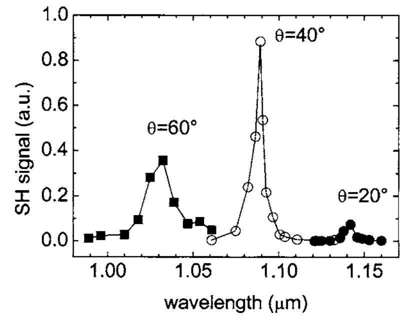

FIG. 3. Experimental results for the transmitted second-harmonic signal as a function of the pump wavelength in the resonance region, for *p*-polarized pump and harmonic waves, at three different values of the incidence angle.

The microcavity has been designed in order to achieve double resonance at a finite incidence angle of 40 ° for *p*-polarized pump and harmonic waves. Exact double resonance condition can be obtained by means of angle tuning: at a given "optimal" angle the low wavelength peak will lie at half the resonance wavelength of the structure. Measurement of transmittance spectra was performed in the 400–2000 nm range by means of a spectrophotometer for different angles and polarizations. The results agree with numerical calculations of the transmission spectrum based on a linear transfer matrix method, in which the dispersive refractive indices measured in Ref. 12 were used for the different layers. In Fig. 2 we show the experimental transmittance for *p*-polarized impinging light at 40 ° (open dots) in a wavelength range centered on the fundamental resonance (1.09 μm) and the transmission at the doubled frequency (full dots). It is to be noted that the transmission peaks overlap: in other words, double resonance occurs. The lines are the results of the numerical calculation. We emphasize that the double resonance condition is obtained with periodic DBR, yielding an improvement on the approach of Ref. 3 where the double resonance was obtained with nonperiodic mirrors. As a matter of fact, in the case of periodic DBR the resonant features are robust with respect to moderate deviations of the layer thicknesses. Such deviations (plausible in realistic growth performed by molecular beam epitaxy or PECVD) produce at most a resonance shift, which can be compensated by tuning the angle of incidence and the polarizations of input and output beams.

Measurements of reflected and transmitted second harmonic signals as a function of both wavelength and polarization were performed in the spectral range 0.9–1.25 μm by means of the idler beam of an optical parametric oscillator pumped by a frequency tripled *Q*-switched Nd:yttrium–aluminum–garnet laser (6 ns time duration laser pulses). The laser beam polarization direction was controlled by means of a double Fresnel rhomb. The transmitted and reflected beams at the fundamental wavelength were absorbed by a glass filter. The reflected and transmitted second harmonic (SH) passed through an analysis polarizer and, therefore, all the possible polarization configurations for the fundamental and SH beam could be investigated. Finally, the SH signals were collected by a pair of monochromators equipped with photomultipliers. The spectral responsivities of the monochromators and photomultipliers were experimentally determined by means of a certified calibration lamp in order to correct the SH signal spectra.

At first instance, only *p*-polarized SH signal was detected. Moreover, the SH signal intensity did not change by rotating the sample around an axis parallel to the growth direction. These results indicate that the second harmonic was generated by an isotropic distribution of nonlinear dipoles, as expected in the case of amorphous materials. Large SH harmonic signals at the fundamental resonance were detected, while lower intensity signals were obtained as the fundamental beam was tuned in the stop band region. Other features appeared in correspondence to the band edges, in agreement with other experimental observations on microcavity systems.[13]

It is well known that SH generation from centrosymmetric materials can result from two different sources: surface nonlinearity and nonlocal bulk quadrupole nonlinearity. In Ref. 14 a clear evidence was found of two resonances at about 1.2 and 1.4 eV, in the SHG spectrum, independently of the polarization state of the impinging beam. These results strongly suggest that the (local) surface contribution in second order nonlinear susceptibility is the dominant one in amorphous silicon, as those spectral resonances are close to the 1.15 and 1.3 eV resonances observed by Pedersen and Morgen in the SHG spectrum of bulk silicon[15] due to transitions from rest-atom to ad-atom dangling-bond surface states. The same conclusion is expected to apply also to the a-$Si_{1-x}N_x$:H multilayers investigated here. In order to verify this, we monitored the SH signal of a single nitride layer while pouring it in methanol. The SH signal was quenched by about 30% of its value by dipping the layer surface. The SH quenching was found to be completely reversible, as after few minutes the SH signal came back to its initial value once the sample was exposed again to air. The same behavior was observed by using acetone. This is a strong indication of the fact that the second order nonlinearity is mainly due to surface $\chi^{(2)}$. This information is important in order to establish a proper theoretical model for interpreting experimental results, as discussed above.

In order to show the effect of the double resonance conditions on the SH emission efficiency, different scans of the SH signal around the resonant wavelength were performed at different angles of incidence. In Fig. 3 we report, on the same scale, three different SH spectra obtained at angles of incidence 20, 40 and 60°. It is noticed that the signal is much more intense at 40 °, which corresponds to the best condition for double resonance.

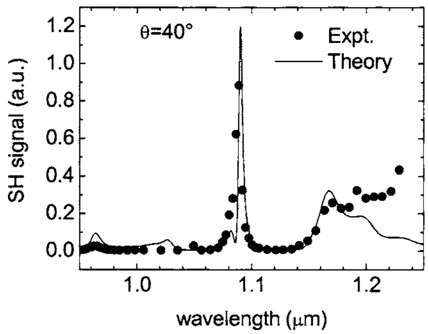

FIG. 4. Transmitted second-harmonic signal as a function of the pump wavelength, for *p*-polarized pump and harmonic waves, at an incidence angle of 40°.

In Fig. 4 we show a typical SH spectrum obtained in transmission geometry for a *p*-polarized fundamental beam. The full dots are the experimental data, while the continuous line is the result of the numerical calculation which we will discuss below. The SH signal increases by two orders of magnitude as the fundamental wavelength goes from the first order stop band to a resonant wavelength of the structure (central resonance and band-edge peaks): the central peak in the SH spectrum corresponds to the double resonance of the microcavity, while the other features correspond to the edges of the stop bands. The resonant enhancement can be ascribed to the high intensity of the light beams at the fundamental and harmonic frequencies at the interfaces surrounding the cavity layer. It is interesting to note that the SH signal detected at the central resonance was hundreds of times larger than the one obtained on a single amorphous silicon nitride film.

We notice also that, due to the "surface" rather than "bulk" nature of the effect, the analytical expression for the SHG efficiency given elsewhere,[6] which applies for a noncentrosymmetric spacer embedded between two mirrors, cannot be invoked to apply in this case. Therefore, a direct comparison of the harmonic efficiency between the doubly resonant microcavity and a single resonance microcavity made by the same materials[10] can hardly be made in our case. In order to model our experimental results, the transmitted SH intensity was calculated by means of the nonlinear transfer matrix method.[11] The surface $\chi^{(2)}$ was described by introducing very thin layers of thickness δ (typically 0.1 nm) at the interfaces, with a refractive index which is the average between those at the two sides of the interface, and an effective $\chi^{(2)}_{\rm bulk}=\chi^{(2)}/\delta$. The surface nonlinear susceptibility has three nonvanishing components: $\chi^{(2)}_{zzz}$, $\chi^{(2)}_{xzx}$ and $\chi^{(2)}_{zxx}$. Since the surface $\chi^{(2)}$ is generated from dipoles which have a component pointing from the low to the high refractive index medium (see Fig. 1, lower panel), each of the $\chi^{(2)}$ components changes its sign when the ordering of the refractive indices is reversed.[16,17] For simplicity, we assumed the surface $\chi^{(2)}$ of the a-$Si_{0.57}N_{0.43}/a$-Si_3N_4 and a-Si_3N_4/a-$Si_{0.45}N_{0.55}$ interfaces to be the same. Notwithstanding this approximation, good agreement with the experimental results is obtained. Good correspondence between theory and experiment is found also for spectra acquired in reflection geometry and with different polarization configurations.

In conclusion, we report the realization of a doubly resonant microcavity based on periodic mirrors and its second harmonic emission properties. We show that the double resonance condition significantly enhances the second harmonic conversion efficiency. We also demonstrate that the experimental results are adequately described by means of surface second order nonlinearity, as confirmed by the good agreement with the theoretical calculation based on nonlinear polarization localized at the interfaces. The results open the way towards improving the conversion efficiency in monolithic cavities made of centrosymmetric or amorphous materials.

[1] A. Ashkin and G. D. Boyd, IEEE J. Quantum Electron. **QE-2**, 109 (1966).
[2] Z. Y. Ou and H. J. Kimble, Opt. Lett. **18**, 1053 (1993).
[3] C. Simonneau, J. P. Debray, J. C. Harmand, P. Vidakovic, D. J. Lovering, and J. A. Levenson, Opt. Lett. **22**, 1775 (1997).
[4] V. Berger, J. Opt. Soc. Am. B **14**, 1351 (1997).
[5] M. Centini, C. Sibilia, M. Scalora, G. D'Aguanno, M. Bertolotti, M. J. Bloemer, C. M. Bowden, and I. Nefedov, Phys. Rev. E **60**, 4891 (1999).
[6] M. Liscidini and L. C. Andreani, Appl. Phys. Lett. **85**, 1883 (2004).
[7] F. F. Ren, R. Li, C. Cheng, H. T. Wang, J. Qiu, J. Si, and K. Hirao, Phys. Rev. B **70**, 245109 (2004).
[8] F. Giorgis, Appl. Phys. Lett. **77**, 522 (2000).
[9] V. Ballarini, G. Barucca, E. Bennici, C. F. Pirri, C. Ricciardi, E. Tresso, and F. Giorgis, Physica E (Amsterdam) **16**, 591 (2003).
[10] S. Lettieri, S. Di Finizio, P. Maddalena, V. Ballarini, and F. Giorgis, Appl. Phys. Lett. **81**, 4706 (2002).
[11] D. S. Bethune, J. Opt. Soc. Am. B **6**, 910 (1989).
[12] C. Ricciardi, V. Ballarini, F. Giorgis, M. Losurdo, G. Bruno, M. Galli, F. Marabelli, M. Liscidini, S. Lettieri, F. Gesuele, and P. Maddalena (unpublished).
[13] T. V. Dolgova, A. I. Maidykovski, and M. G. Martemyanov, Appl. Phys. Lett. **81**, 2725 (2002).
[14] W. M. M. Kessels, J. J. H. Gielis, I. M. P. Aarts, C. M. Leewis, and M. C. M. van de Sanden, Appl. Phys. Lett. **85**, 4049 (2004).
[15] K. Pedersen and F. Morgen, Phys. Rev. B **52**, R2277 (1995).
[16] A. Feigel, Z. Kotler, and B. Sfez, Phys. Rev. B **65**, 073105 (2002).
[17] The sign change of $\chi^{(2)}$ (or of the effective $\chi^{(2)}_{\rm bulk}$) when the interface is reversed is analogous to the mechanism occurring in periodic poling, which may be used to achieve quasi phase matching in isotropic nonlinear materials.