Large interband second-order susceptibilities in $In_xGa_{1-x}N/GaN$ quantum wells

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We present measurements of second-harmonic generation in interband transitions of $In_xGa_{1-x}N/GaN$ multiple quantum well samples. The second-order susceptibility $\chi^{(2)}$ is studied as a function of pump wavelength and quantum well width. For the narrowest wells, we obtain $\chi^{(2)} = 1.3 \pm 0.4 \times 10^{-10}$ m/V, which is an order of magnitude larger than the intrinsic value for bulk GaN. The corresponding power conversion efficiency was 6.3×10^{-7}. An enhancement of the nonlinearity due to strong internal piezoelectric fields could not be observed. © *1999 American Institute of Physics.* [S0003-6951(99)04049-8]

Optical devices based on gallium nitrides have received continuously growing interest over the last few years. The driving force behind these research efforts is the realization of highly efficient light sources in the green, blue, and ultraviolet region of the optical spectrum which has led to the development of light-emitting diodes, in-plane, and surface-emitting lasers.[1] Surprisingly, the nonlinear optical properties of nitrides have received very little attention so far. Miragliotta *et al.* have experimentally determined the bulk value of the second-order susceptibility $\chi^{(2)}$ in bulk GaN (Ref. 2) and have found increased nonlinearities when a large dc electric field is applied to the surface of a GaN film.[3] However, no measurements of the second-order nonlinearity of interband transitions in nitride-based quantum well samples have been carried out. This is all the more surprising since linear optical measurements demonstrated the existence of large internal piezoelectric fields in such quantum wells,[4,5] which are expected to have a significant effect on the nonlinear properties as well.[6]

In this letter, we present measurements of the second-order susceptibility for *interband* transitions in $In_xGa_{1-x}N/GaN$ multiple quantum well (MQW) samples by means of second-harmonic generation (SHG). The SHG intensity was investigated as a function of pump wavelength and quantum well width both to determine absolute values for the nonlinearity and to identify the role of the large internal piezoelectric fields. In addition, we discuss a couple of potential applications for these interband nonlinearities.

It is well known that second-order nonlinear effects can only be observed in noncentrosymmetric structures. In two-dimensional semiconductor samples, this requires asymmetric quantum wells (AQW). These are commonly realized by applying an external electric field[7] or by using compositionally asymmetric coupled quantum wells.[8] In nitride-based quantum wells, however, the intrinsically present piezoelectric field breaks the inversion symmetry automatically. A second effect of the piezoelectric field is the addition of a third-order nonlinear process to the generated second-harmonic (SH) polarization. This effect is called electric-field-induced SH generation (EFISH).[9] Hence, in the presence of a dc piezoelectric field, the nonlinear signal contains two contributions:

$$P(2\omega) = \epsilon_0[\chi^{(2)}(-2\omega,\omega,\omega)E(\omega)E(\omega) + \chi^{(3)}(-2\omega,\omega,\omega,0)E(\omega)E(\omega)E_z(0)], \qquad (1)$$

where $P(2\omega)$ is the generated nonlinear polarization, $E(\omega)$ is the electric field of the fundamental wave, and E_z is the dc piezoelectric field in the growth direction of the sample which we denote as the z direction.

For our experiment, we used a series of MQW samples with 14 periods of undoped $In_{0.13}Ga_{0.87}N$ quantum wells separated by moderately doped (Si, 2×10^{18} cm^{-3}) GaN barriers (43 Å thickness). The quantum well thickness L varied between 12 and 50 Å and other details of the sample structure and growth conditions are described elsewhere.[10] Due to the small total thickness of the quantum well layers, phase matching is not an issue. The samples were optically pumped with pulses (pulse width ≈100 fs) from a mode-locked Ti:sapphire laser at wavelengths between 760 and 890 nm. The pump light was focused to a spot size of ≈10 μm diam on the sample and the typical average excitation powers were around 100 mW. The generated second harmonic was recollimated and the residual pump light was filtered out. For spectrally resolved detection, the second harmonic was collected with a SPEX double-grating monochromator and recorded with a standard photon counting system. Integrated power measurements were taken by focusing the SH light onto a calibrated Si detector and using lock-in detection. All measurements were carried out at room temperature. The pump light was linearly polarized along the $[1\bar{1}00]$ direction of the sample under an angle of 20° with respect to the surface normal. As a consequence, the pump electric field contains x, y, and z components. The point group of the III nitrides is 6 mm and the second-order tensor $\chi^{(2)}$ has seven components. The relevant EFISH third-order tensor $\chi^{(3)}$ has seven nonzero components as well for a dc field in the z direction. We cannot distinguish the $\chi^{(2)}$ and $\chi^{(3)}$ contributions by variation of the incident polarization. Instead, an external variation in the piezofield is necessary, which was

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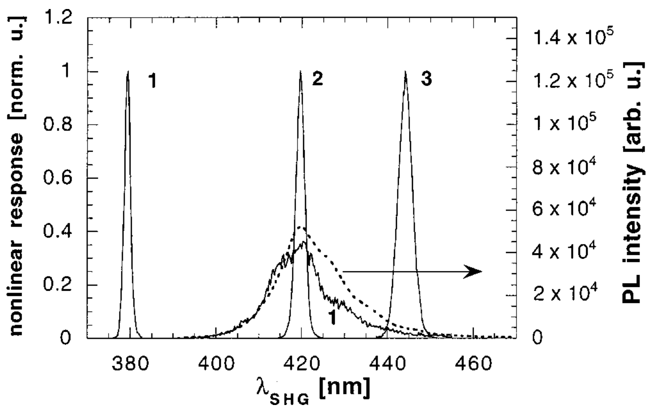

FIG. 1. Cw photoluminescence after excitation in the barriers (dashed line) and nonlinear luminescence spectra (curves 1–3) of the $In_{0.13}Ga_{0.87}N/GaN$ MQW sample with $L=36$ Å. Curves 1–3 were measured for excitation at 760, 840, and 890 nm, respectively, and show narrow resonances due to second-harmonic generation. For SHG wavelengths above the QW transition (curve 1), additional broad luminescence due to two-photon absorption is observed.

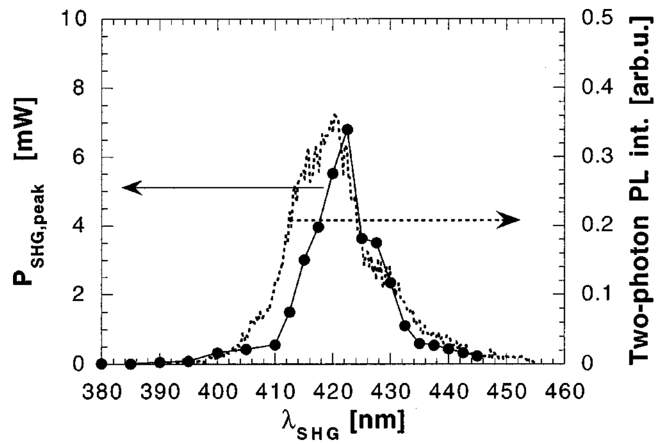

FIG. 2. Spectral dependence of second-harmonic peak power (circles) and two-photon luminescence (dashed line) for $L=36$ Å. The nonlinear response shows a resonance corresponding to the luminescence peak.

not possible in our samples. Since the generated nonlinear polarization also contains components in all three crystal directions, we measure an effective second-order nonlinearity $\chi^{(2)}_{\mathrm{eff}}=:\chi^{(2)}$ (Ref. 11) in our experiment.

In Fig. 1, we show the (normalized) nonlinear spectral response of the sample ($L=36$ Å) for three different pump wavelengths along with the photoluminescence (PL) spectrum obtained from excitation far above the band gap (dashed line). For a pump wavelength whose second harmonic lies well above the first quantum well (QW) transition (curve 1), we observe two distinct features in the spectrum. On one hand, we find a very narrow peak at exactly half of the pump wavelength, and on the other hand, there is a broad peak which coincides with the linear PL spectrum. This broad structure originates from two-photon fluorescence by real carriers which were generated by two-photon absorption. In contrast, the narrow SHG peak is due to virtual carriers from the parametric conversion process. As we increase the pump wavelength towards SHG at the PL resonance, the relative contribution of the SHG signal increases and for resonant excitation (curve 2) we only see the narrow SHG peak. Likewise, for detunings below the quantum well transition (curve 3), we still find a measurable SHG signal but no fluorescence due to the absence of two-photon absorption.

In order to get a quantitative measure of the nonlinearity, we recorded the generated SHG power as a function of SHG wavelength, which is shown in Fig. 2 (circles) along with the two-photon luminescence spectrum from the same sample spot (dotted line). The SHG power spectrum shows a similar resonance as the luminescence in agreement with theoretical expectations. This behavior was observed for all samples. The average SHG power on resonance is $\overline{P}_{2\omega}=60$ nW for an average pump power of $\overline{P}_{\omega}=100$ mW, which corresponds to a power conversion efficiency of 6×10^{-7}. The generated *peak* SHG power $\hat{P}_{2\omega}$ is related to the *peak* pump power $\hat{P}_{\omega}$ by[11]

$$\hat{P}_{2\omega}=\frac{512\pi^4|\chi^{(2)}|^2L^2\hat{P}_{\omega}^2}{n_{\omega}^2 n_{2\omega} d^2 c\lambda^2}, \tag{2}$$

where L is the total nonlinear interaction length, n_{ω}, $n_{2\omega}$ are the refractive indices at the pump and SHG frequency, respectively, d is the excitation spot diameter, c is the vacuum speed of light, and λ is the pump wavelength. We verified the predicted quadratic dependence of SHG on input power by varying the average pump power between 5 and 200 mW and found excellent agreement. This ensures that the SHG has not reached saturation for a pump power of 100 mW.

Using Eq. (2) and the experimentally determined values for peak input and SHG powers, we calculated the value for the second-order susceptibilities in our samples. We also calibrated our setup by measuring the nonlinearity of a commercial KD*P crystal and found good agreement between the measured and theoretical $\chi^{(2)}$ value. In Fig. 3, we depict the maximum values of the second-order susceptibility as a function of quantum well width. We find a strong increase for $\chi^{(2)}$ with decreasing well width. In particular, for the $L=12$ Å well, the susceptibility is found to be $1.3\pm0.4\times10^{-10}$ m/V. This value is large considering that the SH generation is only singly resonant, which typically leads to small nonlinearities.[12] Our value is an order of magnitude larger than the bulk nonlinearity in GaN (Ref. 2) and also larger than the values found in GaAs/AlGaAs-based samples.[13]

The nonlinearity was also expected to show a strong dependence on well width due to the large internal piezoelec-

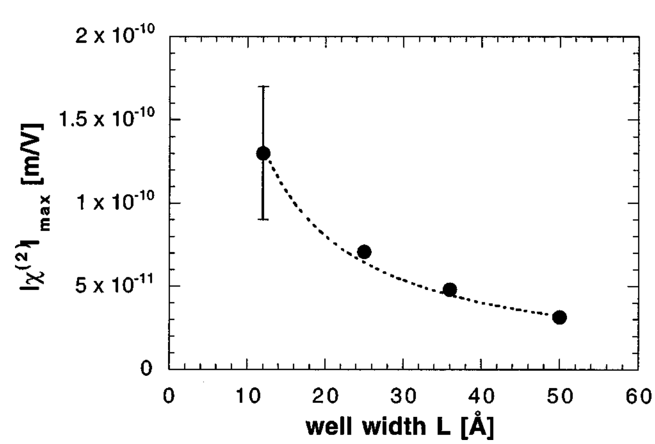

FIG. 3. Maximum second-order susceptibility vs quantum well width L. Larger values are obtained for thinner quantum wells. The dashed line is a fit with an L^{-1} dependence.

tric fields in these samples, which have been estimated to be as high as 1 MV/cm.[4] It has been shown that these fields imply a strong quantum-confined Stark effect, which leads to spatial separation of electron and hole wave functions as the well width increases.[5] Consequently, the oscillator strength and the dipole matrix elements for these transitions are reduced, which should also reduce the optical nonlinearity. Therefore, the nonlinearity should be much smaller for wider quantum wells. The experimentally observed dependence on well width, however, reflects the intrinsic $1/L$ dependence of $\chi^{(2)}$ (Ref. 14) and is well described by a fit using this functional relation (dashed line in Fig. 3). Therefore, we cannot find any indication for an influence of the piezoelectric field from our data, in contrast to theoretical expectations. The reason for this is not clear at this point. On one hand, sample quality generally varies significantly even across the same wafer. The error bar in Fig. 3, for instance, is a result of the variation of $\chi^{(2)}$ across the sample with $L=12$ Å. It is, therefore, not certain how much the quality varies between different samples and how much uncertainty is contained in the well width series. On the other hand, Fiore *et al.* calculated second-order susceptibilities for interband transitions in GaAs/AlGaAs structures and found an almost vanishing dependence of the peak $\chi^{(2)}$ on well width.[15] If there is indeed a weak intrinsic dependence of the susceptibility on well width, then the observed strong increase would be compatible with the presence of large piezoelectric fields. This issue can be clarified by more detailed experiments in which the piezofield strength can be modified externally.

In summary, we have presented measurements of interband second-harmonic generation in nitride-based quantum wells. Large second-order susceptibilities of up to $\chi^{(2)}=1.3\times10^{-10}$ m/V were found. A strong dependence on the well width was observed, which is most likely due to the intrinsic width dependence of $\chi^{(2)}$, and does not appear to be determined by internal piezoelectric fields. Nevertheless, these nonlinearities could have potential applications especially for parametric frequency conversion. For instance, one can use difference frequency conversion to generate visible red light. This could be accomplished in a bottom-emitting vertical-cavity surface-emitting laser structure whose blue emission pumps an underlying nonlinear amplification region which is designed as a resonator for the desired wavelength. Second, due to the large attainable conduction-band offsets in the nitrides, it is also conceivable to generate intersubband emission and parametric amplification at 1.3 or 1.55 μm. This can be realized in a doubly resonant scheme where the pump field is applied between the heavy-hole and second-electronic level and the signal field is tuned to the resonance between the heavy-hole and first-conduction subband, thereby generating an idler field between the two electronic subbands.

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[1] S. Nakamura, M. Senoh, S.-I. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, Y. Sugimoto, and H. Kiyoku, Jpn. J. Appl. Phys., Part 1 **35**, 74 (1996).
[2] J. Miragliotta, W. A. Bryden, T. J. Kistenmacher, and D. K. Wickenden, Mater. Res. Soc. Symp. Proc. **339**, 465 (1994).
[3] J. Miragliotta and D. K. Wickenden, Phys. Rev. B **53**, 1388 (1996).
[4] T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys., Part 2 **36**, L382 (1997).
[5] J. S. Im, H. Kollmer, J. Off, A. Sohmer, F. Scholz, and A. Hangleiter, Phys. Rev. B **57**, R9435 (1998).
[6] C. Bulutay, N. Dagli, and A. Imamoğlu, IEEE J. Quantum Electron. **35**, 590 (1999).
[7] M. M. Fejer, S. J. B. Yoo, R. L. Byer, A. Harwit, and J. S. Harris, Phys. Rev. Lett. **62**, 1041 (1989).
[8] E. Rosencher and P. Bois, Phys. Rev. B **44**, 11315 (1991).
[9] C. H. Lee, R. K. Chang, and N. Bloembergen, Phys. Rev. Lett. **18**, 167 (1967).
[10] S. F. Chichibu, A. C. Abare, M. S. Minsky, S. Keller, S. B. Fleischer, J. E. Bowers, E. Hu, U. K. Mishra, L. A. Coldren, and S. P. Denbaars, Appl. Phys. Lett. **73**, 2006 (1998).
[11] F. Zernike and J. E. Midwinter, *Applied Nonlinear Optics* (Wiley, New York, 1967).
[12] R. Boyd, *Nonlinear Optics* (Academic, Boston, MA, 1992).
[13] A. Fiore, E. Rosencher, V. Berger, and J. Nagle, Appl. Phys. Lett. **67**, 3765 (1995).
[14] R. Atanasov, F. Bassani, and V. M. Agranovich, Phys. Rev. B **50**, 7809 (1994).
[15] A. Fiore, E. Rosencher, B. Vinter, D. Weill, and V. Berger, Phys. Rev. B **51**, 13192 (1995).