Quantum Electronics **46** (10) 888–890 (2016)

NONLINEAR OPTICAL PHENOMENA DOI: 10.1070/QEL16197

Femtosecond superradiance in semiconductor lasers: anomalous internal second-harmonic generation

P.P. Vasil'ev, A.N. Putilin, A.B. Sergeev

Abstract. **The emission of anomalously bright blue light under internal doubling of the frequency of femtosecond superradiance pulses in the active medium of semiconductor GaAs/AlGaAs laser heterostructures has been experimentally found. The efficiency of the internal second-harmonic generation is an order of magnitude higher than in the conventional lasing regime. This effect is due to the formation of a transient ordered state of electrons and holes under superradiance, occurrence of dynamic coherent population lattices, and periodic modulation of the nonlinear susceptibility of the medium.**

Keywords: *superradiance, femtosecond pulses, second-harmonic generation.*

1. Introduction

It is well known that semiconductors, in particular, GaAs or InP and solid solutions on their basis, have pronounced nonlinear optical properties. For example, the nonlinear susceptibility $\chi^{(2)}$ of GaAs is three orders of magnitude higher than that of KDP, a popular nonlinear optical material [1]. One of the sequences of this nonlinearity is second-harmonic generation (SHG) in the active medium of semiconductor lasers. Indeed, even at the dawn of the laser era, it was found that IR GaAs-based lasers emit weak blue light due to the internal SHG [2, 3]. The intensity of this luminescence was proportional to the squared fundamental harmonic power P_ω. It was clear from the very start that, since the absorption of this material at the second-harmonic wavelength is very high, the observed blue light was emitted from a micrometer-sized region near the laser end face. As a result, the conversion efficiency $\eta = P_{2\omega}/P_\omega^2$ turned out to be extremely low (at a level from 10^{-8} to 10^{-10} W^{-1}) [2–5].

At the same time, the superradiance from semiconductor GaAs/AlGaAs laser structures makes it possible to generate femtosecond pulses with an unprecedentedly high (up to 10^9 W cm^{-2}) power at wavelengths of 880–900 nm [6, 7]. Therefore, the giant power of superradiance pulses is expected to make it possible to observe internal second-harmonic radiation of higher intensity than in the conventional lasing regime. Indeed, superradiance experiments revealed a rather intense blue luminescence from the active region of GaAs/AlGaAs laser structures. However, it turned out that the anomalously high intensity of the internal second-harmonic radiation cannot be explained by only an increase in the peak power of superradiance pulses.

In this paper, we report the results of an experimental study of the internal second-harmonic generation under superradiance conditions in GaAs/AlGaAs laser structures. It is shown that the anomalously high second-harmonic intensity may be due to the extraordinary properties of the electron–hole system. Indeed, as was previously found in [8–10], a quantum phase transition under superradiance conditions induces a transient coherent ordered state in the electron–hole system. The condensation of electron–hole pairs in the phase space and the formation of long-range order and dynamic population gratings lead to the occurrence of ideal spatial and temporal coherence and superluminal pulse propagation [11, 12]. We demonstrate below that the same effects may also cause the anomalously strong internal SHG.

2. Experimental

We investigated two types of GaAs/AlGaAs heterostructures with different active-layer configurations. The samples were previously described in detail in [11]. All devices had a three-section geometry and consisted of two amplifying sections at the end faces and a central section (electrically controlled optical absorber). The active GaAs layer (about 0.2 μm thick) was located between p- and n-AlGaAs layers. The samples were divided into two groups: with a standard rectangular active region 5–6 μm wide and with a waveguide expanding from 5 to 40 μm. Nanosecond current pulses with a frequency of 1–10 MHz were applied to the amplifying sections and, depending on the current amplitude, induced electron and hole concentrations in the range of 10^{18}–10^{19} cm^{-3}. An application of a reverse voltage across the central section made it possible to control the optical absorption in the active layer. Spontaneous emission and conventional lasing could be obtained in the structure at different pump levels [9].

In the superradiance regime, the structures emitted pulses with a duration of 300–500 fs, peak power to 120 W, and wavelength in the range of 880–890 nm. Each nanosecond current pulse induced emission of a train consisting of 1 to 12–13 pulses, depending on the reverse voltage across the absorber. The emission dynamics was investigated using a photodetector with a 30-GHz band and a sampling oscilloscope. The pulse durations were measured with a high accuracy using the standard interferometric autocorrelation tech-

P.P. Vasil'ev P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; Centre for Photonic Systems, University of Cambridge, 9 JJ Thomson Avenue, Cambridge, CB3 0FA, UK; e-mail: peter@lebedev.ru;
A.N. Putilin, A.B. Sergeev P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; e-mail: putilinan@sci.lebedev.ru, vnm004@gmail.com

Received 10 August 2016
Kvantovaya Elektronika **46** (10) 888–890 (2016)
Translated by Yu.P. Sin'kov

nique. Two optical filters with a total transmittance of 0.84 and less than 10^{-7} at the wavelengths of the second (440 nm) and first (880 nm) harmonics, respectively, were applied to observe the generation of internal second harmonic and measure its power.

Figure 1 shows a photograph of a collimated second-harmonic beam, recorded by a CCD camera.

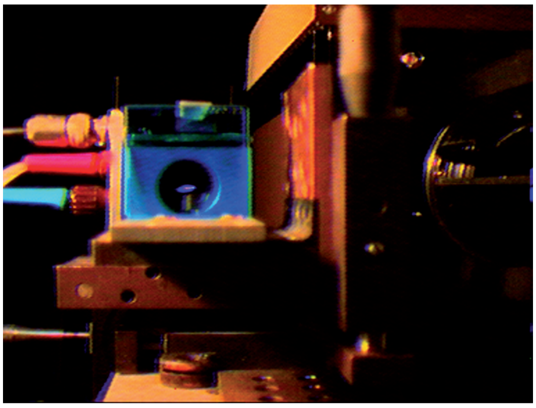

Figure 1. Photograph of the second-harmonic beam.

Emission of blue light was regularly observed in many samples when passing to the superradiance regime; in particular, it was clearly seen in a shaded room on a white screen installed in the beam path. Figure 2 shows a typical dependence of the second-harmonic intensity on the reverse voltage across the absorber in the laser structure.

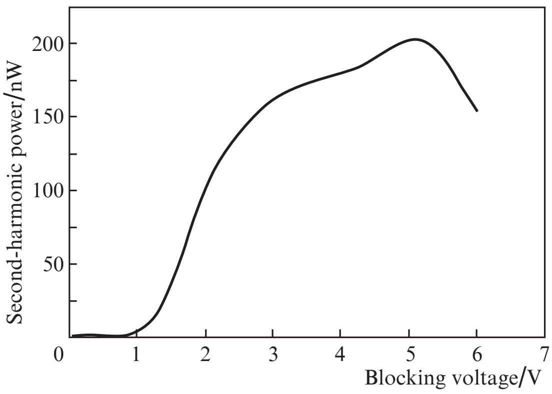

Figure 2. Typical dependence of the average second-harmonic power on the reverse voltage.

At reverse voltages below 1–1.5 V, the conventional lasing regime is implemented. An increase in the voltage leads to the occurrence of superradiance, while the power of femtosecond pulses increases. When the voltage reaches a certain value, the number of superradiance pulses in the train begins to sharply drop, and the second-harmonic intensity decreases (Fig. 2).

The experimental values of the average power of fundamental and second-harmonic radiation and conversion factor η for different samples are listed in Table 1. The experimental data from [3–5] are also given for comparison. In this study, the average superradiance power varied from 1.92 to 9.7 mW (depending on the sample type and pump conditions). The conversion factor η in Table 1 for the samples under study is the ratio of the peak powers of the first and the second harmonics or (the values in parentheses) the ratio of their average powers. The averaged factor η was measured to be 9.0×10^{-7} W^{-1}, a value an order of magnitude larger than that for the conventional lasing mode.

Table 1.

Sample	P_ω	$P_{2\omega}$	η/W^{-1}
From [3]	20 W	10 μW	2.5×10^{-8}
From [4]	30 mW	2.6 pW	2.9×10^{-9}
From [5]	3.4 mW	1 pW	8.0×10^{-8}
H2-51	1.92 mW	193 nW	4.7×10^{-6} (5.2×10^{-2})
H2-31	9.2 mW	104 nW	1.1×10^{-7} (1.2×10^{-3})
H2-33	9.7 mW	130 nW	1.3×10^{-7} (1.4×10^{-3})
H2-52	2.3 mW	36 nW	6.2×10^{-7} (6.8×10^{-3})
H2-63	3.4 mW	43 nW	3.4×10^{-7} (3.7×10^{-3})
H2-112	3.2 mW	41 nW	3.6×10^{-7} (3.9×10^{-3})
H2-131	4.1 mW	56 nW	3.0×10^{-7} (3.3×10^{-3})

3. Results and discussion

The second-harmonic power in the case of internal lasing in the active medium of GaAs/AlGaAs heterostructures can easily be calculated, because the SHG effect has been well studied. The second-harmonic intensity, with a constant first-harmonic power (case of our consideration), is described by the wave equation [13]

$$\frac{\partial E_{2\omega}}{\partial z} = -\frac{\mathrm{i}\omega}{n_{2\omega}c} d_{\mathrm{eff}} E_\omega^2 \mathrm{e}^{i\Delta k z} - \alpha E_{2\omega}, \qquad (1)$$

where d_{eff} is the effective nonlinearity coefficient, Δk is the wave vector mismatch, $n_{2\omega}$ is the refractive index of the medium at the second-harmonic frequency, c is the speed of light, and α is the absorption coefficient at the second-harmonic frequency. Solving Eqn (1), we obtain the second-harmonic power, which is proportional to

$$|E_{2\omega}|^2 \propto \frac{E_\omega^4}{\Delta k^2 + \alpha^2} \{[\cos(\Delta k z) - \mathrm{e}^{-\alpha z}]^2 + [\sin(\Delta k z)]^2\}. \qquad (2)$$

Since $\alpha > 10^7$ m^{-1}, the second-harmonic intensity is saturated at distances on the order of 1 μm. For GaAs, $d_{\mathrm{eff}} = 2.0 \times 10^{-10}$ m V^{-1}, $n_\omega = 3.62$, $n_{2\omega} = 5.04$, and $\alpha = 2.83 \times 10^7$ m^{-1}. Having solved (1) and (2), we obtain the expression for the second-harmonic power flux $I_{2\omega} = c\varepsilon_0 n_{2\omega} |E_{2\omega}|^2/2$ (W m^{-2}):

$$I_{2\omega} = \frac{8\pi^2 d_{\mathrm{eff}}^2}{n_{2\omega} n_\omega^2 c \lambda_\omega^2 \varepsilon_0 (\Delta k^2 + \alpha^2)} I_\omega^2, \qquad (3)$$

where λ_ω is the fundamental wavelength and ε_0 is the permittivity of free space. The proportionality factor in (3) for GaAs is 1.93×10^{-20} m^2 W^{-1}; therefore, in the case under consideration (the emitting area is 2.5×10^{-12} m^2), $\eta = 0.8 \times 10^{-8}$ W^{-1}, which is an order of magnitude smaller than the experimentally found value.

Currently, the physical nature of the generation of anomalously bright internal second-harmonic radiation in the superradiance regime is not completely clear. In our opinion, one of the most likely mechanisms is the interaction of the fields of both the first and second harmonics with the dynamic coherent population gratings [12]. Indeed, population gratings with a half-wavelength period arise during conventional lasing. However, the modulation amplitude of the electron–hole density in this case is only few percent of the threshold density [14]. In the superradiance regime, the working densities of electron–hole pairs are higher than the threshold lasing density by a factor of 3–4 [6, 7], and their modulation depth approaches 100% [14]. Therefore, the periodic $\lambda/2$ modulation of nonlinear susceptibility $\chi^{(2)}$ in the lasing regime is insignificant.

In contrast, when superradiance pulses are generated, coherent population gratings significantly affect the optical field propagation through a semiconductor medium [12].

At the same time, it is well known that variations in the electron–hole density in a semiconductor significantly change its refractive index. Therefore, a spatial population grating in a semiconductor corresponds to a periodic modulation of its refractive index and, correspondingly, a transient spatial grating of its nonlinear susceptibility. Even in the beginning of the 1970s, it was found [15] that a periodic modulation of $\chi^{(2)}$ leads to an increase in the SHG efficiency in comparison with the efficiency for a spatially homogeneous medium. Subsequent experimental and theoretical studies confirmed that the SHG efficiency greatly increases in periodic structures formed in various nonlinear materials (including semiconductors), optical fibers with Bragg gratings, photonic crystals, etc. [16–18]. Note that even a weak periodic modulation of the refractive index of the medium ($\Delta n \sim 10^{-2}-10^{-3}$) may increase the SHG efficiency by more than an order of magnitude [19]. In the case of superradiance in GaAs/AlGaAs heterostructures, as was found previously [12, 14], dynamic coherent population gratings play an important role in the superradiance dynamics, resulting in a considerable decrease in the effective group refractive index of the semiconductor, superluminal propagation of femtosecond pulses, and ultrafast oscillations of the polarisation of the medium. Therefore, it is reasonable to suggest that the periodic modulation of the nonlinear susceptibility of the medium, caused by population gratings, leads to a significant (experimentally observed) increase in the internal SHG efficiency. The theoretical study of this effect is the subject of our next publication.

4. Conclusions

The internal SHG in the superradiance regime in GaAs/AlGaAs heterostructures was investigated. Anomalously bright blue luminescence was revealed in all samples at the transition to the superradiance regime. The conversion efficiency of the superradiance pulse energy to the second harmonic was found to be an order of magnitude higher than in the conventional lasing regime. This effect is explained by the interaction of the optical field of pulses with transient coherent population gratings. The condensation of electron–hole pairs in the phase space at the transition to the superradiance regime [8, 9] results in the formation of an ordered coherent electron–hole state with a deep $\lambda/2$ modulation of the carrier density and corresponding modulation of the refractive index and nonlinear susceptibility. This periodic modulation, as was noted previously, may significantly increase the second-harmonic conversion efficiency.

Acknowledgements. We are grateful to H. Kan and H. Ota for supplying samples of GaAs/AlGaAs heterostructures and to I.V. Smetanin for numerous fruitful discussions.

References

1. *Handbook of Laser Technology and Applications*. Webb C.E., Jones J.D.C. (Eds) (Abingdon: Taylor & Francis, 2004).
2. Garfinkel M., Engeler W.E. *Appl. Phys. Lett*., **3**, 178 (1963).
3. Malmstrom L.D., Schlickman J.J., Kingston R.H. *J. Appl. Phys*., **35**, 248 (1964).
4. Ogasawara N., Ito R., Rokukawa H., Katsurashima W. *Jap. J. Appl. Phys*., **26**, 1386 (1987).
5. Kanamori H., Takashima S., Sakurai K. *Appl. Opt*., **30**, 3795 (1991).
6. Vasil'ev P.P. *Kvantovaya Elektron*., **24**, 885 (1997) [*Quantum Electron*., **27**, 860 (1997)].
7. Vasil'ev P.P. *Kvantovaya Elektron*., **29** (1), 4 (1999) [*Quantum Electron*., **29** (10), 842 (1999)].
8. Vasil'ev P.P. *Phys. Stat. Sol. (b)*, **241**, 1251 (2004).
9. Vasil'ev P.P. *Rep. Progr. Phys*., **72**, 076501 (2009).
10. Vasil'ev P.P., Olle V., Penty R.V., White I.H. *Europhys. Lett*., **104**, 40003 (2013).
11. Vasil'ev P.P., Penty R.V., White I.H. *Kvantovaya Elektron*., **42**, 1081 (2012) [*Quantum Electron*., **42**, 1081 (2012)].
12. Vasil'ev P.P., Penty R.V., White I.H. *Light: Sci. Applications*, **5**, e16086 (2016).
13. Dmitriev V.G., Tarasov L.V. *Prikladnaya nelineinaya optika* (Applied Nonlinear Optics) (Moscow: Fizmatlit, 2004).
14. Vasil'ev P.P., Penty R.V., White I.H. *25th International Semiconductor Laser Conf*. (Kobe, Japan, 2016).
15. Blombergen N., Sievers A.J. *Appl. Phys. Lett*., **17**, 483 (1970).
16. Nakagawa S., Yamada N., Mikoshiba N., Mars D.E. *Appl. Phys. Lett*., **66**, 2159 (1995).
17. Scalora M., Bloemer M.J., Manka A.S., Dowling J.P., Bowden C.M., Vishwanathan R., Haus J.W. *Phys. Rev. A*, **56**, 3166 (1997).
18. Bertolotti M. *J. Opt. A*, **8**, S9 (2006).
19. Haus J.W., Vishwanathan R., Scalora M., Kalocsai A.G., Cole J.D., Theimer J. *Phys. Rev. A*, **57**, 2120 (1998).