Appl. Phys. B 76, 613–616 (2003)
DOI: 10.1007/s00340-003-1167-x

Applied Physics B
Lasers and Optics

Rapid communication

R.KH. ZHUKAVIN[1]
D.M. GAPONOVA[1]
A.V. MURAVJOV[1]
E.E. ORLOVA[1]
V.N. SHASTIN[1]
S.G. PAVLOV[2,✉,*]
H.-W. HÜBERS[2]
J.N. HOVENIER[3]
T.O. KLAASSEN[3]
H. RIEMANN[4]
A.F.G. VAN DER MEER[5]

Laser transitions under resonant optical pumping of donor centres in Si:P

[1] Institute for Physics of Microstructures, Russian Academy of Sciences, GSP-105, 603950 Nizhny Novgorod, Russia
[2] Institut für Weltraumsensorik und Planetenerkundung, DLR, Rutherfordstr. 2, 12489 Berlin, Germany
[3] Delft University of Technology, P.O. Box 5046, 2600 GA Delft, The Netherlands
[4] Institut für Kristallzüchtung, Max-Born-Str. 2, 12489 Berlin, Germany
[5] FOM-Institute for Plasma Physics, Rijnhuizen, 3439 MN Nieuwegein, The Netherlands

Received: 7 November 2002/Revised version: 7 April 2003
Published online: 14 May 2003 •

ABSTRACT Terahertz stimulated emission of phosphorus donors in silicon optically excited by radiation from the free-electron laser FELIX has been studied. It is found that a spectral line of the Si:P laser emission depends on pump frequency. Stimulated emission arises on the $2p_0 \rightarrow 1s(E)$ intra-centre transition (21.2 meV) under resonant pumping of the $2p_0$ state and on the $2p_0 \rightarrow 1s(T_2)$ transition (22.3 meV) under pumping of the $2p_\pm$ or higher odd-parity donor states. The line shift is attributed to the Auger redistribution of the $1s(E)$- and $1s(T_2)$-state populations.

PACS 78.45.+h; 41.60.Cr

1 Introduction

Stimulated emission of shallow impurity centres in semiconductors offers a new and exciting area of research. For the first time optical gain on the intra-centre transitions of gallium (Ga) acceptors has been found in the p-Ge hot hole inter-sub-band laser [1]. Later, THz lasing on the optical transitions between the resonant and bound states of Ga acceptors in germanium under external stress has been reported [2]. Recently, stimulated emission from the phosphorus (P) and bismuth (Bi) group-V donor centres embedded in monocrystalline silicon has been obtained. Donor photoionization by CO_2-laser radiation [3, 4] and resonant pumping of the odd-parity donor excited states by the free-electron laser FELIX [5] were used to achieve Si:P and Si:Bi lasing. Silicon donor lasers operate in the spectral range 5–6 THz [6], which is not bridged by p-Ge hot hole lasers [7] and remains inaccessible for advanced quantum cascade GaAs-based lasers [8, 9].

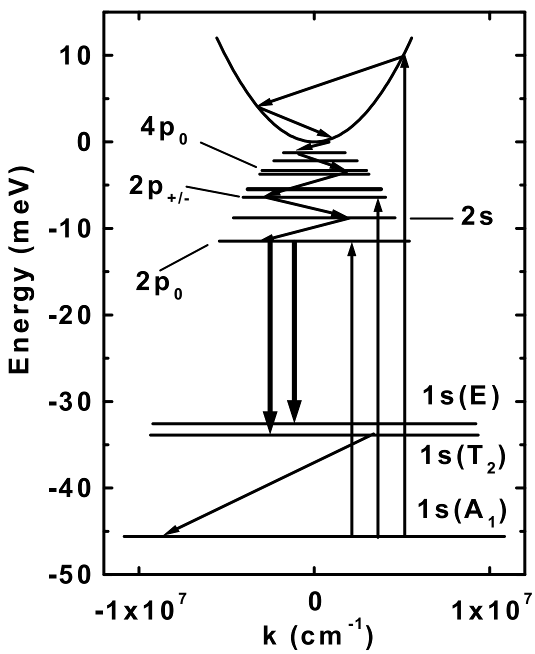

FIGURE 1 Phosphorus donor state optical and nonradiative transitions under conditions of the intra-centre and the photoionizing pumping. Laser transition (*bold arrow*), FELIX pumping (*up arrows*), acoustic phonon-assisted relaxation (*diagonal arrows*)

The mechanism of the population inversion and light amplification on the intra-centre donor transitions in Si:P is based on accumulation of excited carriers on the long-lived $2p_0$ state [10, 11] (Fig. 1).

At a low lattice temperature ($T < 20$ K) the lifetime of excited impurity states is controlled by the acoustic phonon spontaneous emission, whose rates decrease with an increase of the energy gap ΔE between participating bound states. The $2p_0$ state of the P donor is markedly separated ($\Delta E = 21.1$ and $\Delta E = 22.4$ meV) from the lower-lying doublet $1s(E)$ and triplet

✉ Fax: +49-30/6705-55-07, E-mail: sergeij.pavlov@dlr.de
*On leave from: The Institute for Physics of Microstructures, Nizhny Novgorod, Russia

$1s(T_2)$ states and, as a result, the majority of the excited carriers are trapped on the $2p_0$ state due to the intra-centre successive step-by-step relaxation. Calculations made in the frame of the multi-valley model [12] show that the inter-valley phonon-assisted scattering dominates in the relaxation of the $1s(T_2)$ and $2p_0$ states, providing the rates of $3\times10^{10}\,\mathrm{s}^{-1}$ and $2\times10^{9}\,\mathrm{s}^{-1}$ respectively. For the $1s(E)$ state the inter- and the intra-valley scattering processes make comparable contributions to the relaxation rate, which is about $5\times10^{10}\,\mathrm{s}^{-1}$. It should be mentioned that a single-valley approximation yields relaxation rates of $10^{8}\,\mathrm{s}^{-1}$ for the $2p_0$ state and $10^{10}\,\mathrm{s}^{-1}$ for the $1s(E)$ state, while the phonon-assisted scattering from the $1s(T_2)$ state to the nearest $1s(E)$ and $1s(A_1)$ states is forbidden [13]. Different relaxation rates of $2.5\times10^{10}\,\mathrm{s}^{-1}$ for the $1s(E)$ state and $5\times10^{9}\,\mathrm{s}^{-1}$ for the $1s(T_2)$ state were also reported for phosphorus donor centres in Si [14]. Thus, a relatively slow relaxation from the $2p_0$ state leads to population inversion and light amplification on the $2p_0\rightarrow1s(E)$ and the $2p_0\rightarrow1s(T_2)$ transitions [3, 10].

A spectral study of the stimulated emission from Si:P under resonant pumping of odd-parity donor excited states by the free-electron laser FELIX has been conducted in order to gain a better understanding of the intra-centre energy relaxation and to identify laser transitions of donors.

2 Experimental

An experimental Si:P sample (no. 1) was cut in the form of a $7\times5\times1\,\mathrm{mm}^3$ rectangular parallelepiped from a floating zone grown Si crystal and doped by neutron transmutation (see [15]) to donor (P) concentration $N_D=3\times10^{15}\,\mathrm{cm}^{-3}$ and a compensation level $N_A/N_D\approx35\%$. This compensation level was determined by the initial presence of boron acceptors in the Si ingot.

The facets of the sample were set in parallel to each other with an accuracy of 1 arcmin and optically polished to form an internal reflection mode cavity. The sample was cooled to 5 K in a continuous-flow cryostat (Fig. 2) equipped with KRS-5 windows for pump radiation. Cold sapphire and room-temperature polyethylene windows were used for the Si:P laser output.

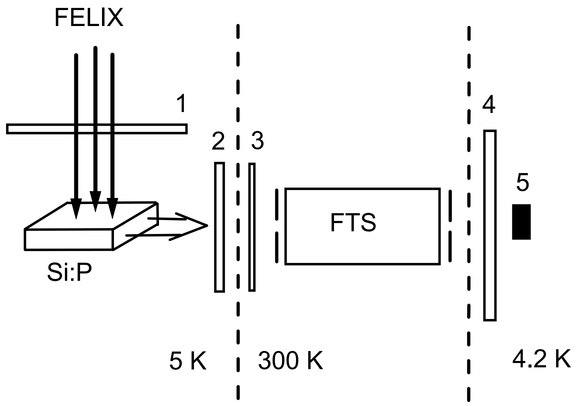

FIGURE 2 The layout of the experimental setup. 1, KRS-5; 2, sapphire; 3, polyethylene windows; 4, CaF_2 filter; 5, Ge:Ga photodetector; FTS, Fourier-transform spectrometer; Si:P, active sample

The free-electron laser FELIX tunable in a wavelength range 25–37 μm emitted 6-μs-long macropulses of far-infrared radiation with a 5-Hz repetition rate. Each macropulse consisted of a train of ∼ 6-ps micropulses with 1-ns time intervals. The micropulse peak power was ∼ 0.5 MW, which corresponded to an average power $P\approx2.5\,\mathrm{kW}$ for a macropulse. The output emission from the Si:P crystal was registered by a liquid-helium (LHe)-cooled Ge:Ga photodetector with a maximum sensitivity in the wavelength range 50–120 μm and with a 10-ns response time. The pump radiation background was filtered from the useful signal by a LHe-cooled 1-mm-thick CaF_2 window. In order to estimate free-carrier concentration in the conduction band, the photocurrent measurements were made on sample no. 2 (dimensions $7\times6\times5\,\mathrm{mm}^3$, $N_D=9\times10^{14}\,\mathrm{cm}^{-3}$, $N_A/N_D\approx1\%$) with ohmic contacts evaporated on the $7\times5\text{-mm}^2$ lateral sides. The FELIX radiation with a $1/e^2$ beam diameter of 12 mm was delivered to the $7\times5\text{-mm}^2$ and $7\times6\text{-mm}^2$ facets of sample nos. 1 and 2 respectively. The incident power was controlled by an external calibrated step-attenuator and measured with a Molectron Energy Max 500 Joule meter installed in front of the flow cryostat. The emission spectra of the Si:P crystal were recorded by a step-scan Fourier-transform spectrometer (FTS) with a spectral resolution of about 0.2 meV.

Figure 3 demonstrates a threshold dependence of THz emission from sample no. 1 on pump intensity under direct excitation of the $2p_0$ donor state, and the temporal behaviour exhibits the rise time of the process. This threshold dependence and also the fact that the emission is observed only from Si:P samples with optically polished facets provide

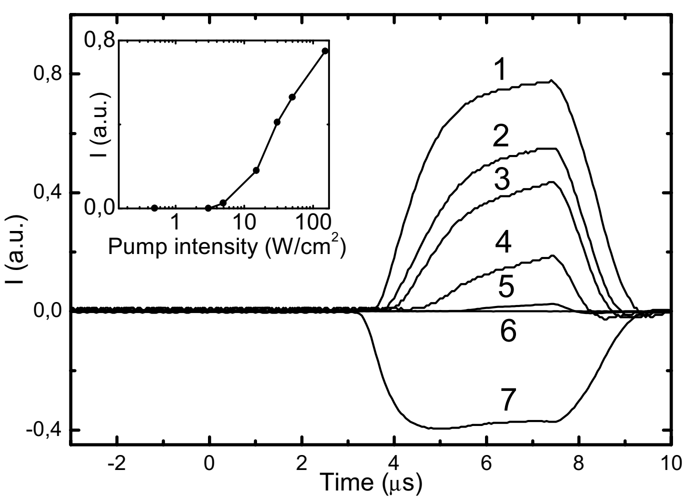

FIGURE 3 Temporal behaviour of stimulated emission from sample no. 1 under direct pumping of the $2p_0$ state for different pump intensities I: (1) 150; (2) 50; (3) 30; (4) 15; (5) 5; (6) 3.5 W cm^{-2}. FELIX macropulse is shown negative (not to scale). *Inset*: Si output versus pump intensity

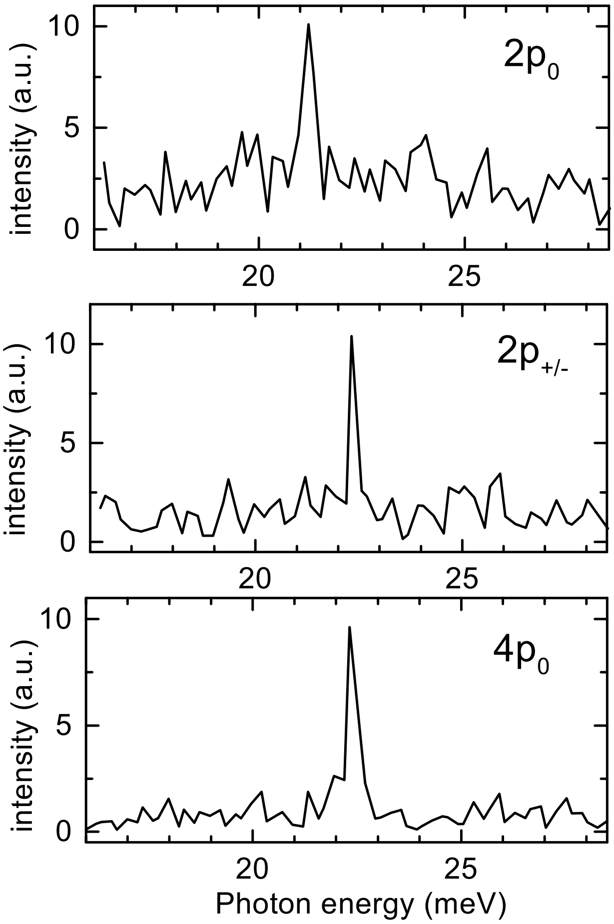

FIGURE 4 Spectra of Si:P stimulated emission (sample no. 1) under pumping in $2p_0$, $2p_\pm$ and $4p_0$ states. Spectral resolution is ~ 0.2 meV. Pump intensity is $\sim 1\,\mathrm{kW\,cm^{-2}}$

evidence of the stimulated character of the emission.

A decrease in the pump power causes a longer delay of the emission pulse with a simultaneous decrease of the output signal. The emission abruptly ceases at the pump power attenuation level of 23 dB, which corresponds to the threshold $I \approx 5\,\mathrm{W\,cm^{-2}}$ (pump intensity averaged over a macropulse), i.e. the photon flux density of 10^{21} photons $\mathrm{cm^{-2}\,s^{-1}}$. Pump power estimates were done taking into account the reflection losses at the cryostat windows and Si crystal.

The emission spectra from Si:P obtained for several wavelengths of FELIX excitation are shown in Fig. 4.

For all wavelengths of the FELIX pump radiation except for 36.4 μm (34 meV, 275 $\mathrm{cm^{-1}}$) the stimulated emission from Si:P was observed on the $2p_0 \to 1s(T_2)$ donor transition at 22.3 meV. However, for the pumping on the wavelength of 36.4 μm, which corresponds to the direct resonant excitation of the $2p_0$ state, stimulated emission occurred on the $2p_0 \to 1s(E)$ transition at 21.2 meV. The same effect was also observed on sample no. 3 with the dimensions $7 \times 7 \times 5\,\mathrm{mm^3}$, $N_D = 3 \times 10^{15}\,\mathrm{cm^{-3}}$ and $N_A/N_D \sim 1\%$.

3 Discussion

The obtained experimental results confirm the basic idea of the theoretical prediction of the effect of stimulated emission on intra-centre transitions of shallow donors in Si [3, 10, 11]. The presented data are in agreement with the earlier results obtained under photoexcitation by FELIX (25–37 μm) as well as by CO_2-laser (9.6–10.6 μm) radiation [3–6]. The threshold dependence of the Si:P output signal (Fig. 3) on pump intensity, which has been observed under direct pumping of odd-parity ($2p_0$, $2p_\pm$, $3p_0$, etc.) donor states as well as continuum states [5], is a weighty argument in witness of the laser action. Net small-signal gain α estimated from the rise time of the stimulated emission increases proportionally to the pump intensity up to $I \approx 50\,\mathrm{W\,cm^{-2}}$, being $\alpha \approx 10^{-3}\,\mathrm{cm^{-1}}$ at the threshold level $I \approx 5\,\mathrm{W\,cm^{-2}}$ (flux density $F \approx 10^{21}$ photons $\mathrm{cm^{-2}\,s^{-1}}$). The saturation of the output signal is observed at the pump intensity $I > 50\,\mathrm{W\,cm^{-2}}$. Unfortunately, we are unable to obtain detailed information of the gain dependence on the pump power by the rise-time measurements because of a slow front of the FELIX macropulse and saturation of the Ge:Ga detector response. The system parameters such as the $2p_0$-state lifetime $\tau_{2p_0} \approx 10^{-9}$ s [12], the amplification cross section $\sigma(2p_0 \to 1s(E)) \approx 5 \times 10^{-15}\,\mathrm{cm^2}$ on the $2p_0 \to 1s(E)$ laser transition [16], the absorption cross section on the transition to the conduction band $\sigma(2p_0 \to \mathit{continuum}) \approx 3 \times 10^{-15}\,\mathrm{cm^2}$ and the cavity losses experimental estimate of $\alpha_c \approx 10^{-2}\,\mathrm{cm^{-1}}$ give the best fit to the experimental results at $I < 50\,\mathrm{W\,cm^{-2}}$. Note that the effective optical cross section $\sigma(1s(A_1) \to 2p_0) \approx 8 \times 10^{-15}\,\mathrm{cm^2}$ and the rate of FELIX pumping ($F \times \sigma$) are reduced by broadening of the FELIX spectral line due to a picosecond duration of the micropulse ($\Delta\hbar\omega \approx 0.2$ meV).

The spectral study (Fig. 4) demonstrates that laser emission from P donors arises either on the $2p_0 \to 1s(E)$ (21.2 meV) or $2p_0 \to 1s(T_2)$ (22.3 meV) lines. Lasing on the $2p_0 \to 1s(E)$ line occurs only under direct pumping of the $2p_0$ state and has never been observed under a CO_2-laser excitation. Selection of one or another line is explained by the laser mode competition process. Lasing on the line with a lower small-signal gain is efficiently suppressed due to depletion of the upper laser state ($2p_0$). The value of the gain $\alpha_{i\to j} = N_D \sigma_{i\to j} \Delta F_{ij}$ depends on the difference in populations, $\Delta F_{ij} = F_i - F_j$, of the principal states and the cross section $\sigma_{i\to j}$ of the radiative transition. The $2p_0 \to 1s(T_2)$ optical cross section is about 1.5 times larger than that for the $2p_0 \to 1s(E)$ transition because of the different degeneracy of the triplet $1s(T_2)$ and the doublet $1s(E)$ [16]. However, the population of the triplet should be higher assuming that it is controlled by the acoustic phonon spontaneous emission. The fact of lasing on the $2p_0 \to 1s(E)$ line (Fig. 4) fits the situation of acoustic phonon-assisted relaxation, but quantitatively it is essential that the lifetime of the $1s(T_2)$ state should be longer than 4×10^{-10} s if the lifetime of the $2p_0$ state is 10^{-9} s. The shift of the

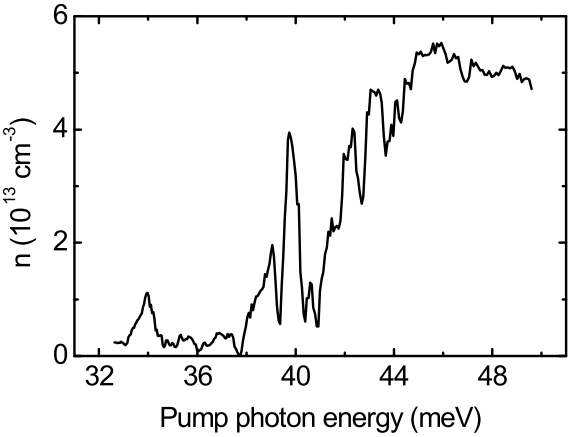

FIGURE 5 Dependence of free-carrier concentration n on pump photon energy for sample no. 2 at pump intensity of 2 kW cm^{-2}. Free-carrier mobility was taken equal to $\sim 3\times10^4$ cm^2 V^{-1} s^{-1}

laser frequency to the $2p_0 \rightarrow 1s(T_2)$ line means that an extra mechanism of the captured carrier redistribution over the bound states is activated with an increase of the pump frequency. We assume that this mechanism is the excitation of donor centres by electron collisions, which tend to equalise the populations of the $1s(E)$ and $1s(T_2)$ states. The Auger process should be effective at providing the cross section of 3×10^{-11} cm^2 and the exchange rate as fast as 10^{-9} s^{-1} at a free-carrier concentration $n \sim 10^{-13}$ cm^{-3} (electron temperature $\sim$ 10–15 K) due to a small energy gap between the levels (1.3 meV). Estimation of the rate with which carriers are transferred from the $1s(T_2)$ state to the $1s(E)$ state under the impact process can be made using a second-order approach of the perturbation theory and will be discussed elsewhere. It is very important to note that photocurrent measurements (Fig. 5) show a steady rise of free-electron concentration with a pump-frequency increase.

In conclusion, the spectra of the stimulated emission from P donor centres in silicon under resonant pumping of different odd-parity excited donor states have been studied. It was found out that direct pumping of the $2p_0$ state changes the spectral line of the stimulated emission. The effect of the line shift is explained by the influence of the Auger process on the $1s(T_2)$- and $1s(E)$-state populations.

ACKNOWLEDGEMENTS This work was partly supported by the Deutsche Forschungsgemeinschaft and the Russian Foundation for Basic Research (RFBR) (Joint Grant Nos. 436 RUS 113/206/0 (R) and 03-02-04010), RFBR Grant Nos. 02-02-16790 and 03-02-16586 and NATO Grant No. PST.EV.978669. S.G. Pavlov gratefully acknowledges support through the Alexander von Humboldt Stiftung.

REFERENCES

1 A.V. Murav'ev, S.G. Pavlov, V.N. Shastin: JETP Lett. **52**, 343 (1990)
2 I.V. Altukhov, M.S. Kagan, K.A. Korolev, V.P. Sinis, E.G. Chirkova, M.A. Odnoblyudov, I.N. Yassievich: JETP **88**, 51 (1999)
3 S.G. Pavlov, R.K. Zhukavin, E.E. Orlova, V.N. Shastin, A.V. Kirsanov, H.-W. Hübers, K. Auen, H. Riemann: Phys. Rev. Lett. **84**, 5220 (2000)
4 H.-W. Hübers, S.G. Pavlov, M. Rümmeli, R.K. Zhukavin, E.E. Orlova, H. Riemann, V.N. Shastin: Physica B **308**, 232 (2001)
5 V.N. Shastin, R.K. Zhukavin, E.E. Orlova, S.G. Pavlov, M.H. Rümmeli, H.-W. Hübers, J.N. Hovenier, T.O. Klaassen, I.V. Bradley, A.F.G. van der Meer: Appl. Phys. Lett. **80**, 3512 (2002)
6 H.-W. Hübers, S.G. Pavlov, M. Greiner-Bär, M.H. Rümmeli, M. Kimmitt, R.K. Zhukavin, H. Riemann, V.N. Shastin: Phys. Status Solidi B **233**, 191 (2002)
7 V.N. Shastin: Opt. Quantum Electron. 23, S111 (1991)
8 R. Köhler, A. Tredicucci, F. Beltram, H.E. Beere, E.H. Linfield, A.G. Davies, D.A. Ritchie, R.C. Iotti, F. Rossi: Nature (Lond.) **417**, 156 (2002)
9 B.S. Williams, H. Callebaut, S. Kumar, Q. Hu, J.L. Reno: Appl. Phys. Lett. **82**, 1015 (2003)
10 V.N. Shastin: in Proc. 21st Int. Conf. Infrared Millimeter Waves, Berlin, 1996, CT2
11 E.E. Orlova, R.K. Zhukavin, S.G. Pavlov, V.N. Shastin: Phys. Status Solidi B **210**, 859 (1998)
12 E.E. Orlova: Ph.D. thesis, Institute for Physics of Microstructures, Nizhny Novgorod, 2002, unpublished
13 A. Griffin, P. Carruthers: Phys. Rev. **131**, 1976 (1963)
14 T.G. Castner: Phys. Rev. **130**, 58 (1963)
15 I.M. Gres'kov, B.V. Smirnov, S.P. Sobolev, A.A. Stuk, V.A. Kharchenko: Sov. Phys. Semicond. **12**, 1118 (1978)
16 A.J. Mayur, M.D. Sciacca, A.K. Ramdas, S. Rodriguez: Phys. Rev. B **48**, 10 893 (1993)