Stimulated terahertz emission from group-V donors in silicon under intracenter photoexcitation

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Frequency-tunable radiation from the free electron laser FELIX was used to excite neutral phosphorus and bismuth donors embedded in bulk monocrystalline silicon. Lasing at terahertz frequencies has been observed at liquid helium temperature while resonant pumping of odd parity impurity states. The threshold was about two orders of magnitude below the value for photoionization pumping. The influence of nonequilibrium intervalley TO phonons on the population of excited Bi impurity states is discussed. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1476955]

Semiconductors doped by shallow level Coulomb centers are promising media for terahertz (THz) light amplification and stimulated emission. Up to the moment the main activity has been concentrated on phosphorus (P) and bismuth (Bi) donors embedded in single crystalline silicon (Si) excited by CO_2 laser radiation. Both, THz spontaneous and stimulated emission based on intracenter optical transitions, were detected and the involved states have been identified.[1–5]

There are two basic mechanisms that may cause the population inversion of charge carriers between the impurity states. The first one is based on the suppression of the acoustical phonons assisted relaxation of the optically excited electrons over the localized states with the increase of the energy gap between the levels. Such a bottleneck effect occurs for the lower excited states of the impurity center. In Si:P it leads to the overpopulation of the $2p_0$ state and THz stimulated emission on the $2p_0 \rightarrow 1s(T)$ transition under optical pumping at cryogenic temperatures ($T < 15\,\mathrm{K}$) [Fig. 1(a)]. A similar effect can be expected for Si doped by As, Sb, Li shallow donors as well. Another mechanism for population inversion is predicted in Si:Bi [Fig. 1(b)] due to the strong coupling of both $2p_0$ and $2s$ excited states with the $1s(A)$ ground state via intervalley TO and LO optical phonon resonant interaction.[6] Spontaneous emission of optical phonons makes the lifetimes of the $2p_0$ and $2s$ states extremely short (10^{-12} s), dumping carriers directly to the ground state. Such a scenario provides the depletion of the $2p_0$ and $2s$ states and leads to a negligible population of the $1s(E,T)$ states. Hence, the population inversion between the higher excited states and the $2s$, $2p_0$, $1s(E,T)$ states is expected.[1,5] Recently THz lasing has been obtained from the $2p_\pm \rightarrow 1s(E,T)$ transitions in Si:Bi under CO_2 laser pumping.[5] The drawbacks of the CO_2 laser pumping, which

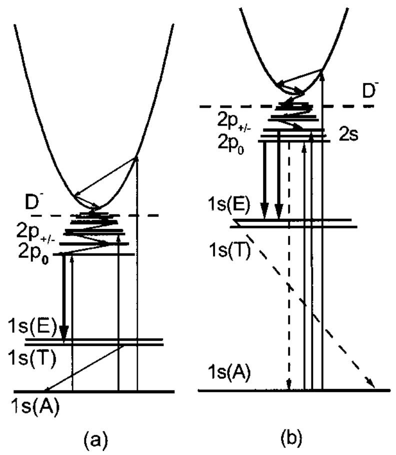

FIG. 1. Scheme of optical and nonradiative transitions in Si:P (a) and Si:Bi (b) under intracenter excitation and photoionization pumping: broad arrow down—THz emission; arrow up—FELIX pumping; diagonal solid arrows—acoustical assisted transition; diagonal dashed arrows—low probable acoustical assisted transitions; dashed vertical arrow down—optical phonon assisted transition.

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TABLE I. The parameters of the Si samples.

Si sample	Dominant impurity	Net doping concentration (cm^{-3})	Sample geometry $l\times b\times c$ (mm^3)	Compensation, N_A/N_D	Doping procedure
1	P	3×10^{15}	$7\times7\times5$	<0.01	During crystal growth
2	P	3×10^{15}	$7\times5\times1$	0.35	Neutron transmutation doping
3	P	3×10^{15}	$7\times5\times1$	<0.001	Neutron transmutation doping
4	Bi	10^{16}	$7\times7\times5$	0.1	During crystal growth
5	P	9×10^{14}	$7\times6\times5$	<0.01	During crystal growth
6	Bi	10^{15}	$7\times7\times2$	⋯	During crystal growth

in fact leads to photoionization (ground impurity state-to-continuum excitation) is twofold. First of all the absorption cross section is small ($\sim4\times10^{-16}$ cm^2 for Si:P)[3] compared to that of intracenter—i.e., between impurity localized states—optical transitions (as high as about 10^{-13} cm^2).[7] Second, the photoionization leads to the creation of D^- centers (neutral donor with an extra electron) which are good absorbers of THz radiation.[8] Both factors increase the pump threshold for stimulated emission.[3,4]

The aim of the pump experiment at the Dutch Free Electron Laser Experiment, FELIX, is to compare the efficiency of intracenter and photoionization pumping for the generation of laser emission and to investigate the intracenter relaxation of photoexcited carriers. Tunable mid- and far-infrared radiation from FELIX gives the unique opportunity to perform the appropriate measurements. The FELIX radiation consisted of 6–8 μs long trains of (6–8 ps long) micropulses at a 1 ns time interval at a repetition rate of 5 Hz. Frequency scans covered donor intracenter absorption bands of 25–36 μm for Si:P and 17–22 μm for Si:Bi. Several silicon samples have been investigated. The most important parameters of the samples are listed in Table I. The samples were shaped in rectangular parallelepipeds with $l\times b\times c$ dimensions and with the facet $l\times b$ perpendicular to the (111) crystal axis. The facets were polished parallel to each other within 1 arc min accuracy forming a mirrorless Fabry–Perot cavity. The refraction index of silicon 3.4 provides the reflection coefficient $R\cong0.3$ for normally incident light. Without the polished cavity stimulated emission was not observed. Investigated Si samples were immersed in liquid helium (LHe). The FELIX radiation with a beam in diameter of 1 cm was guided to the sample $l\times b$ facet by a 70 cm length and 1 cm inner diameter stainless steel light pipe. A step attenuator was used to change the incident power. The photon flux density, averaged over a macropulse was derived from the time averaged FELIX output, as measured with a joulemeter (Energy Max 500, Molectron) in front of the light pipe. The exact power at the position of the crystal is not precisely known, which finally limits the precision of the measurements. THz radiation emitted from the Si crystal $b\times c$ facet was registered by a LHe cooled Ge:Ga detector, with a maximum detectivity in the wavelength range of 50–120 μm. However, this detector still has a finite sensitivity at the shorter wavelengths, and therefore CaF_2 and Al_2O_3 filters have been used to prevent the pump radiation to reach the detector. For alignment of the FELIX beam an additional Ge:Ga photodetector was placed behind the Si crystal. Sample Nos. 5 and 6 with ohmic Sb–Au contacts were used for photocurrent measurements.

Figures 2 and 3 present the dependencies of THz emission from Si:P and Si:Bi samples on the pump photon energy for the different levels of pump power. The threshold character of the emission signal has been found for Si sample Nos. 1–4. The presented data reveal that the most effective pump frequencies correspond to donor intracenter absorption lines, except the case with $1s(A)\rightarrow2p_0$ pumping for Si:Bi.

The lowest value of the threshold pump power density was 50 W/cm^2, measured at the entrance to the light pipe. This has been realized by directly pumping into the $2p_0$ state of Si:P (sample No. 3). The corresponding flux density of 9.2×10^{21}photon$\times$cm$^{-2}\times$s^{-1} is much smaller than the so far lowest observed threshold of 4×10^{23} photon$\times$cm^{-2} $\times$s^{-1} for pumping with a CO_2 laser (pump photon energy is 122 and 117 meV).[3,4] Figures 2(a) and 2(b) allow to compare the laser thresholds of THz emission obtained from Si:P

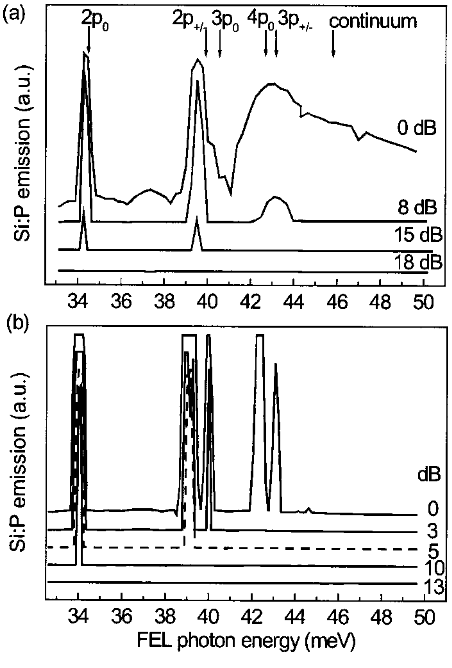

FIG. 2. Dependence of the signal on pump power and frequency for compensated Si sample No. 2(a) and uncompensated Si sample No. 3(b) doped by P. The higher resolution on (b) has been obtained due to the smaller FELIX wavelength step scan. Saturation of the emission from Si with increasing pump intensity is due to the saturation of the Ge:Ga detector.

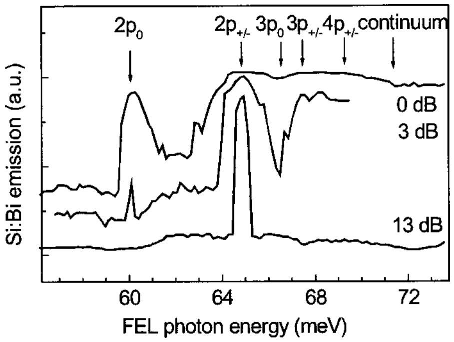

FIG. 3. Dependence of the Si:Bi signal on pump frequency for different pump power levels.

samples with the same doping concentration, but different compensation. Crystal Nos. 2 and 3 [Fig. 2(a) and 2(b), respectively] show a different behavior when the pump photon energy exceeds the photoionization edge (>46 meV). The absence of THz emission in the uncompensated crystal No. 3 might be due to the effect of a larger D^- center concentration and the related THz absorption, which is higher than in the compensated sample. Stimulated emission from Si:P when pumped with maximum power from ground to continuum states ($g.s. \rightarrow \mathrm{cont.}$) was observed only for compensated samples. In addition, the laser threshold pump power density was lower for the compensated crystal ($50\,\mathrm{W} \times \mathrm{cm}^{-2}$) than for the uncompensated crystal ($100\,\mathrm{W} \times \mathrm{cm}^{-2}$). The results of FELIX wavelength scan measurements made for Si:Bi are presented in Fig. 3. Unlike Si:P, stimulated emission from Si:Bi is found to be strongest when pumping into the $2p_\pm$ state (pump photon energy is ~64.6 meV), with the lowest threshold photon flux density 1.6×10^{22} photon$\times \mathrm{cm}^{-2} \times \mathrm{s}^{-1}$ ($\sim 170\,\mathrm{W} \times \mathrm{cm}^{-2}$). In addition, lasing occurs for direct excitation of the $2p_0$ state with a pump photon flux density of 1.8×10^{23} photon$\times \mathrm{cm}^{-2} \times \mathrm{s}^{-1}$ ($1700\,\mathrm{W} \times \mathrm{cm}^{-2}$). This is one order of magnitude higher than for the $2p_\pm$ state.

The photocurrent at a bias voltage of 0.5 V has been measured as a function of pump radiation frequency for Si:P and Si:Bi samples. It did not reveal an essential ionization of donors when pumping the $2p_0$ and $2p_\pm$ states. For Si:P the corresponding photocurrent (estimated concentration of free electrons $n \approx 3 \times 10^{13}\ \mathrm{cm}^{-3}$) at maximum pump flux density was five times less than the photocurrent observed for photoinization pumping. At the laser threshold it was insignificant ($n \approx 3 \times 10^{11}\ \mathrm{cm}^{-3}$). For Si:Bi the photocurrent intracenter excitation was negligible ($n \leqslant 10^{11}\ \mathrm{cm}^{-3}$) even for the maximum of pump power.

To summarize, THz lasing has been observed from P and Bi neutral centers in silicon when optically pumped into the odd parity impurity states. The reduction of the laser threshold by about two orders of magnitude under resonant pumping instead of photoionization by radiation from a CO_2 laser can be explained by the larger absorption cross section for resonant pumping ($2 \times 10^{-14}\ \mathrm{cm}^2$,[7] vs $4 \times 10^{-16}\ \mathrm{cm}^2$ for Si:P). The most effective pumping for Si:P is into the $2p_0$ state. The fact, that the threshold differs not dramatically for pumping into the $2p_\pm$ or in the $3p_0$ excited states, confirms the cascade character of the relaxation through the ladder of excited states. It indicates that excited carriers gradually lose their energy being finally trapped on the most long-living state, i.e., $2p_0$. This is in a good agreement with the theoretical prediction.[1] On the other hand, the measurements on the Si:Bi samples revealed unexpected results. First, the threshold values of photon pump flux density for Si:Bi and Si:P are approximately the same, while the lifetimes of the involved states differ strongly. The estimated lifetime of the $2p_0$ state in Si:P is 1.5×10^{-8} s (Ref. 3) and the lifetime of the $2p_\pm$ state in Si:Bi is about 10^{-9} s.[5] But the most striking fact is the stimulated emission observed for the direct excitation into the $2p_0$ state, which has to have an extremely short time of life due to emission of intervalley TO phonons (10^{-12} s).[6] Additionally, the photocurrent measurements prove that for the $2p_0$ pumping no free carriers are created, so ionization of Bi centers cannot be the cause of a large population of the $2p_\pm$ state that could result in emission. The lasing can be explained by overpopulation of the $2p_0$ state. We suppose that reabsorption of TO phonons by the impurity centers (phonon trapping)[9] produces additional pumping. However, a long (10^{-10} s) lifetime of nonequilibrium intervalley optical phonons is required for such a process.

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