Stimulated THz emission from resonant intra-center optical pumped Si:P and Si:Bi.

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Abstract— *The tunable Free Electron Laser FELIX has been used as resonant intra-center optical pump to study the properties of the stimulated THz emission of P and Bi donors in silicon at liquid helium temperature. Emission spectra and results of time-resolved experiments will be presented and discussed in terms of lifetimes of the impurity states.*

Introduction

SEMICONDUCTORS doped by shallow impurities are promising media for Terahertz (THz) stimulated emission. Up to the moment the main activity has been concentrated on Phosphor (P) and Bismuth (Bi) donors in silicon excited by CO_2 laser radiation. Both spontaneous and stimulated THz emission was detected and the impurity states involved identified [1-4].

There are two basic mechanisms that may cause population inversion of charge carriers between the impurity states. The first one is based on the suppression of acoustic-phonon-assisted relaxation of 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 impurity centers. In Si:P it leads to the overpopulation of the $2p_0$ state and stimulated THz emission on the $2p_0 \rightarrow 1s(E,T_2)$ transitions under optical pumping at cryogenic temperatures (T<15K) (Fig. 1a). A second mechanism for

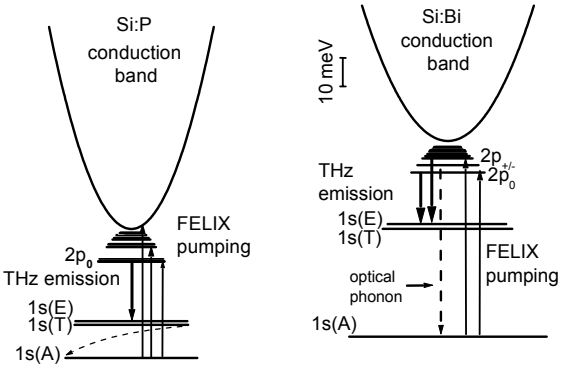

Fig. 1. Energy level schemes of Si:P and Si:Bi

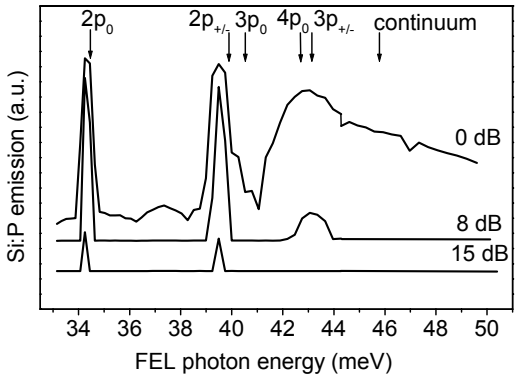

Fig. 2. Si:P Stimulated emission versus frequency at different FELIX pump powers.

population inversion occurs in Si:Bi (Fig. 1b) due to the strong coupling of both the $2p_0$ and 2s states with the 1s(A) ground state via resonant inter-valley TO- and LO- phonon interaction [5]. Spontaneous emission of optical phonons makes the lifetimes of the $2p_0$ and 2s states extremely short (10^{-12} s), dumping carriers directly into the ground state.

This leads to depletion of the $2p_0$ and 2s states and to a negligible population of the $1s(E,T_2)$ states. The resulting population inversion between the higher excited states and the 2s, $2p_0$ and $1s(E,T_2)$ leads to THz emission on the $2p_\pm \rightarrow 1s(E,T_2)$ transitions under CO_2 laser pumping [4]. CO_2 laser pumping leads to photoionization and has some drawbacks. First of all the absorption cross section for impurity groundstate-to-continuum excitation is small, ca. 4×10^{-16} cm^2 for Si:P compared to about 10^{-13} cm^2 for intra-center excitation. Secondly, the photoionization leads to the creation of D^- -centers (neutral donor + electron) which are good absorbers of THz radiation. Both factors increase the pump threshold for stimulated emission [3]. The use of the frequency tunable Dutch Free Electron Laser FELIX as excitation source enables the use of the more efficient intra-center pumping and moreover the investigation of the intra-center relaxation processes of the photoexcited carriers.

Experiments

The FELIX radiation consisted of 6–8 μs long trains of micro-pulses at a repetition rate of 5 Hz. The micropulses, at a 1 ns time interval and a 6–8 ps FWHM have a maximum peak power of about 0.5 MW. The crystals were shaped in rectangular parallelepipeds, forming a mirror-less Fabry–Perot cavity. Either a continous flow cryostat with windows was used, or samples were immersed in liquid Helium, and the radiation was guided to and from the sample by light pipes. Emission was measured with a Ge:Ga detector, and spectra were obtained with a Fourier transform spectrometer (FTS) with a resolution about of 1 cm^{-1}.

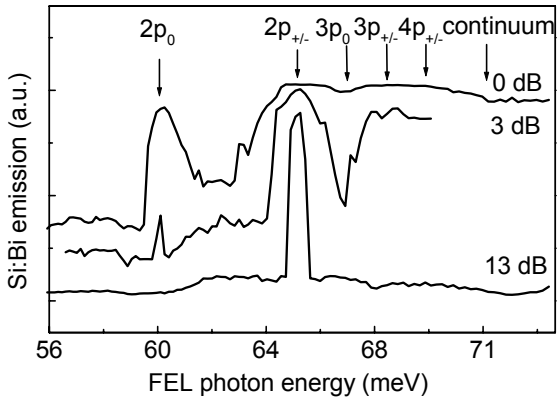

Fig.3. Si:Bi. Stimulated emission as a function of the frequency at different FELIX pumppowers.

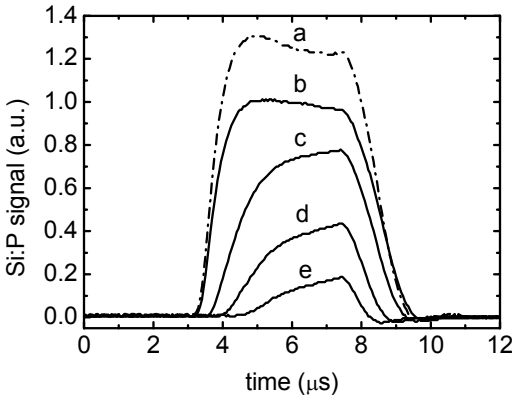

Fig. 4. Pulse shape of the Si:P stimulated emission for different pump intensities under direct excitation of the $2p_0$ state.
(a) FELIX macropulse shape.
Pump power attenuation is 0 dB (b)(micropulse power 160 kW/cm^2), 8 dB(c), 15 dB(d), 18 dB(e)

Results

Figure 2 shows the dependence of the Si:P THz emission on the pump photon energy for different levels of pump power. The presented data reveal that the most effective pump frequency corresponds to the $1s(A)\rightarrow2p_0$ donor intra-center transition. The results obtained for Si:Bi are presented in Fig 3. Unlike Si:P, stimulated emission from Si:Bi is found to be the strongest when exciting the $2p_\pm$ state. In addition, lasing occurs also for

direct excitation of the $2p_0$ state, be it with a threshold pump flux density of about one order of magnitude higher than for the $2p_\pm$ state [6].
The stimulated emission pulses from Si:P while optically exciting the $2p_0$ state are shown in Fig. 4. At the maximum pump power, the shape and duration of the emission pulse is similar to the macropulse envelope of the FELIX radiation. Attenuation of the pump power causes the time delay of the emission pulse to increase to 2.5 μs with a simultaneous decrease of the output signal. The stimulated emission ceases at a pump power attenuation of 23 dB, which corresponds to a pump power averaged over a macropulse of about 5 W/cm^2 , taking into account reflection losses at the windows and at the Si crystal, i.e. a photon flux density of about $1x10^{21}$ photons/cm^{2}s. That is considerably lower than the threshold of 4×10^{23} photons/cm^{2}s, observed earlier for excitation into the conduction band using CO_2 laser radiation [3].
In Fig. 5 spectra from Si:P obtained for different excitation wavelengths are shown. For all pump wavelengths except of 36.4 μm the stimulated emission is observed on the $2p_0 \rightarrow 1s(T_2)$ transition at 180 cm^{-1}. However, for the pump wavelength of 36.4 μm, which corresponds to the excitation from the ground state directly to the $2p_0$ state, stimulated emission occurs on the $2p_0 \rightarrow 1s(E)$ transition at 171 cm^{-1}. This effect is not an artefact of the specific sample, as this emission line switching was also observed for a different samples with different dimensions, donor concentration and compensation [6].
This peculiar behaviour might be related to the presence of two competing factors determining the relative gain of the $2p_0 \rightarrow 1s(E)$ and $2p_0 \rightarrow 1s(T_2)$ transitions. At the one hand, the optical cross section for the optical transition from the $2p_0$ state to the triplet $1s(T_2)$ state is about 1.5 times larger than that to the doublet 1s(E) state. On the other hand, the phonon-assisted relaxation of carriers from the 1s(E) state is faster than that from the $1s(T_2)$ state because only inter-valley phonon-assisted transitions are allowed for the $1s(T_2)$ state, whereas both the intra-valley and the inter-valley phonon-assisted relaxations is allowed for the 1s(E) state. In the case of direct pumping into the $2p_0$ state apparently the $2p_0 \rightarrow 1s(E)$ stimulated emission line dominates due to the lower population of the 1s(E) state resulting from its shorter lifetime. Pumping into the $2p_\pm$ or

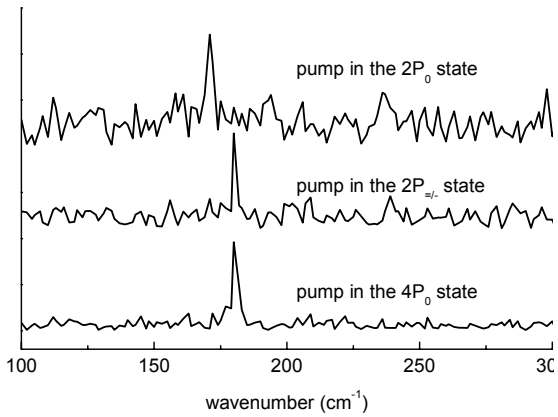

Fig. 5. Si:P: Spectra of the stimulated emission, optically exciting different donor states.

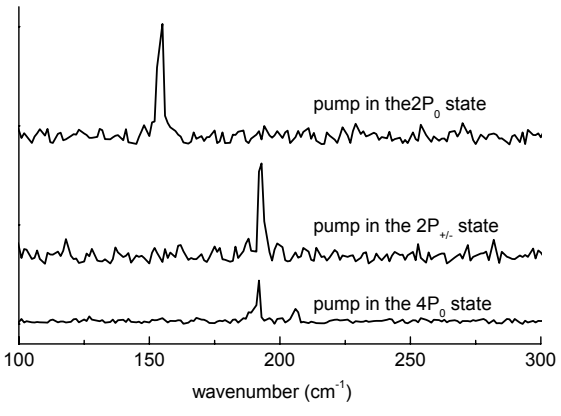

Fig. 6. Si:Bi: Spectra of the stimulated emission, optically exciting different donor states.

higher states the free carrier concentration is about 4 times larger than that for pumping into the $2p_0$ state. Then the exchange of carriers between the 1s(E) and $1s(T_2)$ states caused by Auger processes comes into play. Because of the small energy gap between

these states (~1.3 meV) even at free carriers concentrations about of 10^{13} cm^{-3} the Auger process provides an exchange rate of the order of 10^{10} s^{-1}, which tends to equalize the populations of the 1s(E) and 1s(T_2) states. As a result, the cross section factor becomes dominating, causing the switching of the stimulated emission to the $2p_0 \rightarrow 1s$ (T_2) transition.
A similar behaviour seems to be found in Si:Bi. Now the lowest threshold is observed for pumping the $2p_\pm$ state. Stimulated emission occurs here on the $2p_\pm \rightarrow 1s(E)$ transition at 191 cm^{-1} when exciting the $2p_\pm$ state (Fig.6). Pumping into higher excited donor states (or into the continuum) both the $2p_\pm \rightarrow 1s(E)$ and the $2p_\pm \rightarrow 1s(T_2)$ transitions at 206 cm^{-1} are observed [7]. Possibly the larger ionization energy of the Bi donor causes the free carrier concentration while pumping into the $2p_\pm$ state to be too small for the Auger process to become important.
Surprisingly, under direct optical excitation of the short-living $2p_0$ state, also stimulated emission is observed on the $2p_0 \rightarrow 1s(E)$ transition at 155 cm^{-1}.

Conclusions

Resonant pumping of excited donor states in silicon at cryogenic temperatures leads to population inversion and stimulated THz emission. This intracenter pumping is about two orders of magnitude more efficient than earlier employed photoionization pumping using CO_2 laser radiation.
The spectra of the THz stimulated emission from the phosphor and bismuth donors in silicon, exciting different odd-parity states, have been obtained and the responsible intra-center optical transitions have been determined. A peculiar change of the emission transition has been observed as a function of the particular donor state that is optically excited. Tentatively, this effect is explained by the presence of free carriers which, through an of Auger process, tends to equalize the population of the 1s(E) and 1s(T_2) states involved in the stimulated emission transitions.

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