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Far-infrared stimulated emission from optically excited bismuth donors in silicon

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Far-infrared stimulated emission from optically pumped neutral Bi donors in silicon has been obtained. Lasing with wavelengths of 52.2 and 48.6 μm from the intra-center $2p_{\pm} \rightarrow 1s(E{:}\Gamma_8),1s(T_2{:}\Gamma_8)$ transitions has been realized under CO_2 laser pumping. The population inversion mechanism is based on fast optical-phonon-assisted relaxation from the $2p_0$ and $2s$ excited states directly to the ground $1s(A)$ state leading to relatively small population in the intermediate $1s(E)$, $1s(T_2)$ excited states. © *2002 American Institute of Physics.*
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Coherent radiation sources for the far-infrared (FIR) part of the electromagnetic spectrum are required for applications in astronomy and atmospheric remote sensing as well as molecular and solid-state spectroscopy. There is a lack of tunable fundamental oscillators in this region. Among the known FIR semiconductor lasers are long-wavelength tunable intersubband hot hole p-Ge lasers,[1] mid-infrared narrow band gap lead salt semiconductor lasers[2] as well as intersubband quantum "fountain" [3] and "cascade" lasers.[4] Recently, a FIR silicon laser has been demonstrated. Transitions between optically excited hydrogen-like impurity centers are used to obtain FIR spontaneous and stimulated emission.[5–8]

Here we report on a population inversion mechanism in silicon doped by bismuth (Si:Bi) impurity centers. In contrast to phosphor doped silicon, where population inversion is based on the long-living $2p_0$ state,[7] Si:Bi has no long-living state. The $2p_0$ and $2s$ states are coupled to the $1s(A)$ ground state by a resonant interaction via an optical phonon[9] and have a very short lifetime of about 1 ps. Due to this, the majority of the optically excited electrons relaxes directly to the $1s(A)$ state and, therefore, does not reach the $1s(E)$ and $1s(T_2)$ states. As a result, the $2s$ and $2p_0$ states as well as the valley-orbit splitted $1s(E)$ and $1s(T_2)$ states remain essentially unpopulated. Therefore, population inversion can be achieved between the populated states, higher than the $2s$ state, and the excited states lying below despite the fact that their lifetime is almost the same. Consequently, laser action can occur on the allowed optical transitions, from the lowest of the populated states, $2p_{\pm}$, to the closest unpopulated $1s(E)$ and $1s(T_2)$ states (Fig. 1).

In more detail, the mechanism is as follows: optically excited free carriers lose their energy through interaction with acoustic and optical phonons being further captured by ionized impurity centers. The cascade capture[10] assisted by acoustic phonons means a gradual relaxation of the excited carriers through the ladder of excited impurity states. Transitions between neighboring lower impurity states dominate and the probability $P(i)$ that an excited carrier comes to such a state i before relaxing to the ground state is close to unity.[5] The situation is different for the $1s(E)$ and $1s(T_2)$ states. The rate of transitions from the $2p_0$ and $2s$ states to the ground state due to spontaneous emission of optical phonons is much higher than the rate of acoustic-phonon-assisted transitions from the $2p_0$ to the $1s(E)$ and $1s(T_2)$ states. Carriers are trapped in the $1s(E)$ and $1s(T_2)$ states via

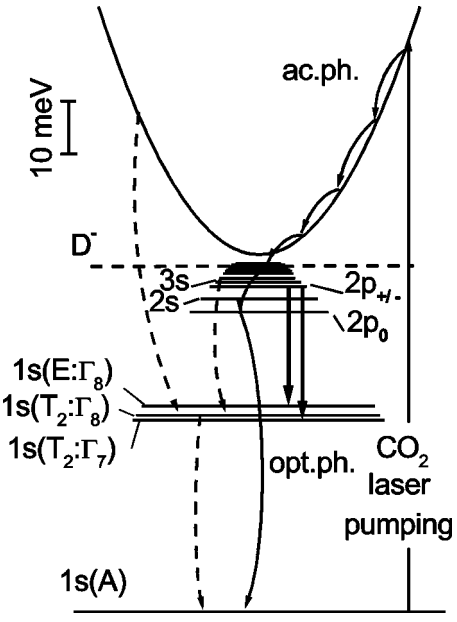

FIG. 1. Scheme of optical and nonradiative transitions in Si:Bi under CO_2 laser pumping: straight arrows down—FIR laser emission; dash curved arrows down—low-probable relaxation of the photoexcited electrons due to emission of acoustic and optical phonons; solid curved arrows down—major relaxation of the electrons due to emission of acoustic and optical phonons. D^- is the D^- center energy level. Quadruplet $1s(T_2{:}\Gamma_8)$ and doublet $1s(T_2{:}\Gamma_7)$ states are spin-orbit splitted $1s(T_2{:}\Gamma_5)$ state (labeled as in Ref. 13).

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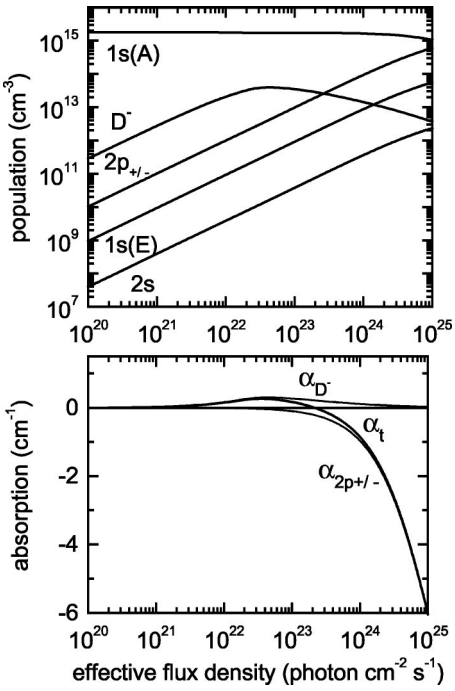

FIG. 2. Calculated population (upper graph) of the ground $1s(A)$ and of the excited $1s(E)$, $2s$, $2p_{\pm}$ states and D^- centers. Absorption (lower graph) by neutral donor and D^- center transitions vs pump flux density for $T=4$ K, Bi concentration 2×10^{15} cm^{-3} and compensation level of 10%. The term α_{D^-} indicates the absorption of 52 μm radiation by D^- centers, $\alpha_{2p+/-}$ is the amplification on the $2p_{\pm}\rightarrow1s(E)$ transitions, taking into count absorption on transitions from the $2p_{\pm}$ state to the conduction band, α_t is the net gain at 52 μm on the $2p_{\pm}\rightarrow1s(E)$ transitions.

optical-phonon-assisted recombination from the conduction band and direct acoustic-phonon-assisted transitions from the $3s$ state. The population of the excited impurity states is given by[5]

$$N_i = N_{\text{g.s.}} W^{\text{opt}} P(i)\tau_i, \quad \text{for } e^{(E_i - E_{\text{g.s.}})/kT} \ll W^{\text{opt}} P(i)\tau_i,$$

where W^{opt} is the optical ionization rate, τ_i and E_i are the lifetime and binding energy of the excited state i, $N_{\text{g.s.}}$ and $E_{\text{g.s.}}$ are the ground state population and ionization energy respectively, and T is the lattice temperature. The probability to populate the $2p_{\pm}$ state due to cascade relaxation, $P(2p_{\pm})$, is calculated to be about 0.5, while for the $1s(E)$ and $1s(T_2)$ states it is $P(1s(E,T))\simeq3\times10^{-3}$. Therefore, although the lifetime of the $2p_{\pm}$ state and the lower $1s(E)$ and $1s(T_2)$ states is almost the same (in the order of 10^{-10} s), the latter are less populated and a four-level laser scheme can be realized from Bi donor transitions.

A model has been developed to assess the laser mechanism quantitatively and to guide the experiments. In Fig. 2 the population of impurity states calculated using a probability technique[5] and amplification coefficients for different pump flux densities are presented. The calculations take into account the absorption of the FIR emission due to the D^- centers[11] (neutral D^0 centers with an extra electron), which appear when the impurity centers are photoionized. The transition rates were estimated assuming the hydrogen-like center model for D^0 centers[12] and the zero-radius potential for D^- centers. The calculations define the optimal Bi concentration to be in the range of 10^{15}–10^{16} cm^{-3}.

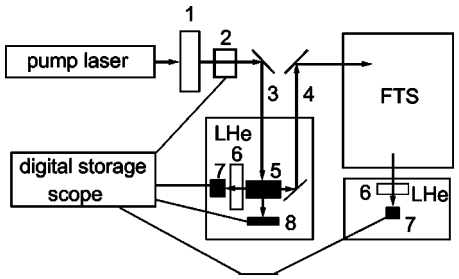

FIG. 3. Experimental setup for observation of emission from Si:Bi: (1) pump laser attenuator, (2) photon drag monitor, (3) pump beam, (4) emission from Si:Bi, (5) Si:Bi sample, (6) FIR filter set, (7) FIR detectors, (8) detector for pump beam alignment.

The investigated Si:Bi samples with donor concentrations 5×10^{13}–1.3×10^{16} cm^{-3} were cut in the form of rectangular parallelepipeds ($7\times7\times5$ mm^3) from the same ingot. The purer crystal ($\sim5\times10^{13}$ cm^{-3}) was used to identify the dominant dopant as well as the concentration of the incorporated electrically active centers using absorption spectroscopy. All absorption lines in these spectra correspond to Bi intra-center optical transitions. The compensation of the Si:Bi samples was determined to be about 10%. The samples were mounted in a holder and immersed in a liquid helium (LHe) vessel. A pulsed TEA (Transverse Excited Atomspheric) CO_2 laser with peak output power up to 2 MW in the wavelength range 9.2–10.7 μm was used to pump the silicon samples (Fig. 3). The effective pump photon flux density inside the silicon sample was derived from the CO_2 laser pulse peak power and taking into account reflections from the optical window, the crystal surface as well as lattice and impurity absorption losses. The total loss of the pump power is estimated to be 7 dB. The FIR emission from the Si:Bi samples was registered by a LHe cooled Ge:Ga photodetector, sensitive in the wavelength range 40–125 μm. To prevent irradiation of the detector by the CO_2 laser, a 1-mm-thick sapphire filter was used.

Spontaneous emission in the wavelength range of the Ge detector was registered from Si:Bi samples with doping levels greater than 5×10^{14} cm^{-3} for various pump wavelengths of the CO_2 laser. A linear dependence of the spontaneous emission signal on the pump photon flux density up to 2×10^{25} cm$^{-2}\times$s^{-1} was found (Fig. 4). The spontaneous emission signal increased by about two times with increasing

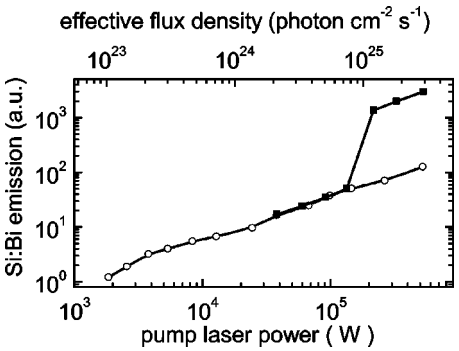

FIG. 4. Dependence of FIR emission at a wavelength of 52.2 μm from Si:Bi on pump power of the CO_2 laser at 10.6 μm (10P20 line). Open circles are for spontaneous emission from the unpolished Si:Bi sample. Solid squares are for stimulated emission from the same Si:Bi sample after optical treatment.

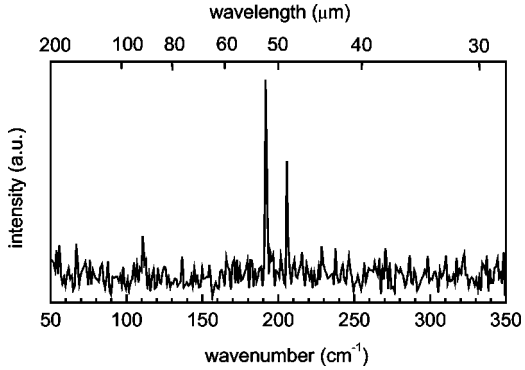

FIG. 5. Stimulated emission spectrum from Si:Bi. The two emission lines are identified as the $2p_{\pm}\rightarrow 1s(E{:}\Gamma_8)$ and $2p_{\pm}\rightarrow 1s(T_2{:}\Gamma_8)$ intra-center Bi transitions.

pump photon energy from 116 to 135 meV (wavelengths 10.7 and 9.2 μm). Stimulated emission has been observed from the samples with a doping concentration in the range of $(2-13)\times 10^{15}$ cm^{-3} which were optically treated to provide a high-Q resonator. The effective threshold photon flux pump density for different samples was $(5-8)\times 10^{24}$ photons $\times$cm$^{-2}\times$s^{-1} (Fig. 4). The laser threshold was a factor 1.5–1.8 lower for the 9 μm pump emission band (photon energy 130–135 meV). Samples with a doping level below 8×10^{14} cm^{-3} did not show stimulated emission. The stimulated emission spectrum for the Si:Bi sample with a doping concentration of 1.3×10^{16} cm^{-3} was measured by the FIR Fourier transform spectrometer with a resolution of 1 cm^{-1} (Fig. 5). The observed lines at 191.6 cm^{-1} (52.2 μm) and 205.6 cm^{-1} (48.6 μm) correspond to the $2p_{\pm}\rightarrow 1s(E{:}\Gamma_8)$ and $2p_{\pm}\rightarrow 1s(T_2{:}\Gamma_8)$ intra-center Bi transitions (labeled according to the notation used in Ref. 13). Both emission lines have the same laser threshold.

The model, described above, is in agreement with the observed laser transitions. However, the measurements revealed a larger laser threshold. This can be explained because the calculations do not take into account Auger processes, which can take place for the free electron concentrations $n\geqslant 10^{14}$ cm^{-3}, when the pumping photon flux density$\geqslant 10^{24}$ cm$^{-2}\times$s^{-1}. The Auger process decreases the lifetime of the $2p_{\pm}$ laser state throwing carriers down into the $2s$ state. This reduces the gain and leads to the larger threshold intensity. In addition, it can explain the pump frequency dependence. Pumping by a line from the 9 μm band of the CO_2 laser provides less free carriers than pumping by a line from the 10 μm band due to the intra-band optical-phonon-assisted recombination.

In summary, we have demonstrated a laser mechanism based on resonant optical phonon interaction of excited states of shallow impurity centers in semiconductors. These results give encouragement to use intra-center transitions for the generation of far-infrared radiation.

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