Stimulated Emission from Donor Transitions in Silicon

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(Received 29 November 1999)

The observation of far-infrared stimulated emission from shallow donor transitions in silicon is reported. Lasing with a wavelength of 59 μm due to the neutral donor intracenter $2p_0 \rightarrow 1s(E)$ transition in Si:P pumped by CO_2 laser radiation is obtained. Populations of D^0 and D^- center states and the balance of the radiation absorption and amplification are theoretically analyzed.

PACS numbers: 78.45.+h, 42.55.Rz, 71.55.–i

Semiconductor sources of stimulated emission which are based on intraband optical transitions of highly nonequilibrium charge carriers are the object of an extensively developing field in physics. The new approaches in the development of unipolar active media are promising to bridge the wavelength range from $\lambda = 1$ μm up to $\lambda = 1000$ μm of the electromagnetic spectrum. Examples of sources based on bulk material are germanium hot hole lasers [1] and Er-doped silicon lasers [2]. Emitters based on the properties of a two-dimensional electron gas in different kinds of heterostructures are quantum well "cascade" [3] and "fountain" lasers [4] as well as miniband Bloch oscillators in superlattices [5].

We propose a new far-infrared (FIR) (55 μm $< \lambda <$ 65 μm) active medium based on neutral donor (D^0) intracenter optical transitions in bulk silicon doped by elements of group V of the periodic table (P, As, Sb). Relatively low threshold pump power for laser operation and high quantum efficiency, caused by the specific character of the intracenter phonon relaxation, are the expected merits of this type of laser. The physical concept, the first observation of stimulated emission from Si:P under excitation by a CO_2 laser and a theoretical model, is presented below.

The mechanism for the intracenter population inversion is based on the accumulation of charge carriers in the long-lived bound excited state of neutral donors. For group V elements in Si this is usually the $2p_0$ state. At low lattice temperature ($T < 30$ K) the interaction with intervalley and optical phonons is negligible for these centers. Therefore, the population of the bound excited states of the donors is controlled by long-wavelength acoustical phonon assisted relaxation.

The matrix elements of acoustical phonon assisted transitions depend crucially on the wave functions of the initial and final states. They decrease with increasing energy gap, ΔE, between the levels of the bound states provided $qa \gg 1$ (a: effective radius of the orbit; q: the wave vector of phonon required for the transition, $\hbar qs = \Delta E$; s: speed of sound) [6]. This corresponds to the case when the phonon wave vector exceeds the localization of the wave function of the impurity state in q space. Consequently, transitions between neighboring levels predominate for lower ($n \leq 3$) bound states. This leads to a step-by-step cascade relaxation. Thus, the majority of excited carriers accumulates in the $2p_0$ state before returning to the ground state.

In Si:P (Fig. 1) the largest energy gap occurs between the $2p_0$ and the $1s(E,T)$ states which are split off by the intervalley interaction. Calculation of the lifetime of the $2p_0$ state leads to a value of $\tau_{2p0} \simeq 1.5 \times 10^{-8}$ s for a temperature of $T = 5$ K. In comparison, the lifetime of the $1s(E)$ state is much shorter, $\tau_{1sE} \simeq 2 \times 10^{-10}$ s, because of the smaller energy gap down to the $1s(A)$ ground state. This is a four level laser scheme (Fig. 1) with a thresholdless population inversion on the $2p_0 \rightarrow 1s(E,T)$ transitions.

The threshold of the stimulated emission is determined by the balance of amplification on the $2p_0 \rightarrow 1s(E,T)$ transitions and the absorption in the crystal. The latter includes the absorption by transitions from bound D^0 states to continuum states (with a cross section $\sigma_D \approx 5 \times 10^{-15}$ cm^2), lattice absorption ($\alpha < 1.0$ cm^{-1} [7]), and cavity losses. In comparison, the amplification cross section of the $2p_0 \rightarrow 1s(E,T)$ transitions is $\sigma_{21} \lesssim 10^{-13}$ cm^2 [8]. This can be deduced from the full width at half maximum $\Delta = 0.4$ cm^{-1} of these lines which was observed in Si:P crystals of highest quality and with a donor concentration $N_D = 2 \times 10^{15}$ cm^{-3}. The amplification cross section decreases with increasing dislocation

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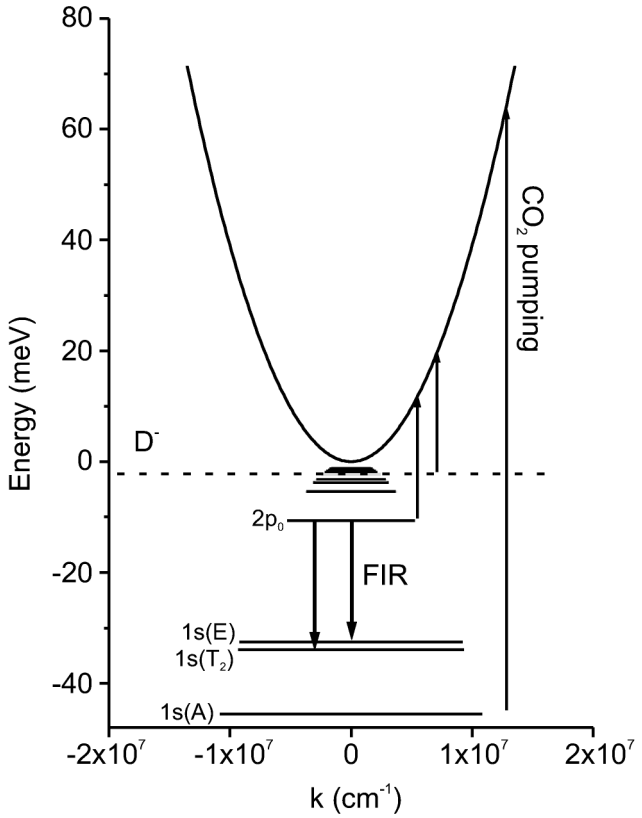

FIG. 1. Optical transitions in Si:P. The dashed line represents the energy level of the D^- center state.

density, doping concentration, and with temperature increase.

Optical excitation with a CO_2 laser provides an effective mechanism to populate the $2p_0$ state of Si:P. By absorption of photons from the CO_2 laser, free electrons are produced in the conduction band. They loose their energy quite fast via optical ($\tau_{\text{opt}} \simeq 10^{-12}$ s) and acoustical ($\tau_{\text{ac}} \simeq 10^{-10}$ s) phonon emission and are further captured by ionized donors with a capture cross section $\sigma_c \simeq 5 \times 10^{-11}$ cm^2 for $T = 5$ K. Optically excited carriers can also be trapped by D^0 centers (cross section $\sigma_c \simeq 10^{-13}$ cm^2) forming negatively charged donor ions D^- which can significantly contribute to the absorption (cross section $\sigma_{D^-} \approx 10^{-14}$ cm^2 [9]) (Fig. 1).

Previous calculations [10] and FIR transmission measurements in Si:P under CO_2 laser pumping [11] allowed us to define the range of doping concentrations which are necessary for the generation of stimulated emission at liquid He temperatures. The investigated samples are listed in Table I.

The residual boron acceptor concentration in all samples was less than 10^{13} cm^{-3}. The samples were cut in the form of rectangular parallelepipeds ($a \times l \times d$ mm^3). The facets $a \times d$ mm^2 at the ends of the longest (l) axis were polished parallel to each other within 1 arcmin accuracy to form a mirrorless (reflection coefficient $\eta \approx 0.3$) Fabry-Perót cavity.

A Q-switched CO_2 laser (peak power up to 500 W) and a TEA CO_2 laser (peak power up to 500 kW) were used. Both operate with an output wavelength $\lambda = 10.6$ μm with pulse durations of 1 and 0.2 μs, respectively. A stainless steel lightpipe guided the pump radiation to the sample under investigation ($a \times l$ mm^2 facet) which was immersed in liquid He. The FIR emission was detected transversely to the pump radiation by a Ge:Ga (Ge40) photodetector which is sensitive in the wavelength range 30 to 120 μm [see inset in Fig. 3 (below)]. The CO_2 laser radiation was prevented from reaching the Ge40 detector by means of a sapphire filter. The emission spectrum was analyzed by placing InSb, GaAs, and KRS-5 filters in front of the Ge40 detector.

The dependence of the far-infrared emission on the CO_2 laser intensity (photon flux density P) is presented in Fig. 2. A linear dependence for spontaneous emission with respect to the CO_2 laser pump power was observed for a CO_2 laser intensity less than 200 W cm^{-2} ($P = 10^{22}$ photons cm^{-2} s^{-1}) for all the samples (see inset in Fig. 2). Stimulated emission was observed for samples 2, 3, and 4 with donor concentrations in the range $N_D = (0.9\text{–}3) \times 10^{15}$ cm^{-3}. The output FIR signal increases by at least 2 orders of magnitude with an optical threshold above 8 kW cm^{-2} ($P \geq 4 \times 10^{23}$ photons cm^{-2} s^{-1}) for the higher doped samples 3 and 4 and above 100 kW cm^{-2} ($P \geq 5 \times 10^{24}$ photons cm^{-2} s^{-1}) for the purer sample 2. The other samples listed in Table I did not show threshold emission enhancement. However, the photocurrent possesses a quiet smooth, close to square root behavior for all the samples. The free carrier concentration does not exceed $n = 10^{14}$ cm^{-3} for the photon flux densities up to 10^{25} photons cm^{-2} s^{-1}. The effect was not observed with the sample of length $l \leq 5$ mm (sample 3a). The pulse of the FIR emission signal (Fig. 3) was shorter than the CO_2 laser radiation pulse with the intensity above threshold, while for the lower radiation intensity the pulses are similar. The inserted filters indicate that the FIR signal originated from the $2p_0 \rightarrow 1s(E)$ transition with a wavelength $\lambda = 59$ μm.

The pronounced threshold increase of the FIR signal, which is not accompanied by a dramatic redistribution of charge carriers, proves the stimulated character of FIR emission for $P \geq 4 \times 10^{23}$ photons cm^{-2} s^{-1}. The shorter duration of the emission pulse above threshold is explained by the exponential development of the emission pulse and its decay due to Si lattice heating during the CO_2 laser pumping pulse. The absence of this effect for the 5 mm length sample (3a, Table I) permits an estimate of the small signal gain (including cavity losses), which yields 2 cm^{-1} for $N_D = 2 \times 10^{15}$ cm^{-3}. Increase of the donor concentration leads to an enhancement of the amplification until concentration broadening of the intracenter levels becomes significant and thus terminates the gain. This explains the concentration dependence of the effect.

TABLE I. The parameters of the samples.

Si sample	Method of growth	Net phosphorus doping concentration (cm^{-3})	Sample geometry $a \times l \times d$ (mm^3)
1	Czochralski	$<6 \times 10^{13}$	$7 \times 7 \times 5$
2	Float zone	9×10^{14}	$7 \times 7 \times 5$
3	Czochralski	2×10^{15}	$7 \times 7 \times 5$
3a	Czochralski	2×10^{15}	$4 \times 5 \times 4$
4	Float zone	3×10^{15}	$7 \times 7 \times 5$
5	Czochralski	9×10^{15}	$7 \times 7 \times 5$

For the analysis of the experimental results the population density of the D^0 and D^- centers as well as free carrier concentration were calculated as a function of the pump power from the CO_2 laser. A temperature of 4.2 K for the Si:P crystal has been assumed throughout the calculations (Fig. 4a). Solving the system of rate equations only for the several lowest bound excited states does not yield sufficiently accurate results because the upper excited states significantly influence the population of the lower excited states. This is due to the fast transition rates between such states. In order to take into account the influence of cascade transitions between excited states on the population of a particular bound excited state, a modification of the method of sticking probabilities introduced by Lax [12] was used. This technique is described elsewhere in more detail [10]. The matrix elements of acoustical phonon assisted transitions between bound excited states were calculated using the single valley approximation [6] (compare [13]) and assumed an isotropic band with an effective mass $m_{\text{eff}} = 0.33 m_0$ (m_0: free electron mass). For a more accurate determination of the lifetime of the $2p_0$ state, calculations of the corresponding matrix elements were made using the method of asymptotic short wavelength [14] and by taking into account the many-valley effect.

On this basis the FIR absorption coefficients were calculated using the parameters of sample 3 (Table I). The linewidth of the $2p_0 \rightarrow 1s(E)$ transition is taken as 2 cm^{-1}. The result of the calculations is presented in Fig. 4b. The calculated amplification coefficient is about 3 times higher and the calculated threshold pumping level is 4 times less than the observed values. This discrepancy can be explained by additional absorption due to $D^- - D^+$ complexes which have not been taken into account in the present treatment.

It is worth noting that the absorption due to D^- centers decreases for a pump intensity $P \geq 10^{23}$ photons $cm^{-2}\,s^{-1}$ because of their photoionization by the pump radiation. The absorption on D^- centers can be reduced in compensated samples. The threshold pump intensity can be minimized to about $P \simeq 2 \times 10^{22}$ photons $cm^{-2}\,s^{-1}$ (see Fig. 4) by using external mirrors with an optimal output power coupling and compensated Si:P.

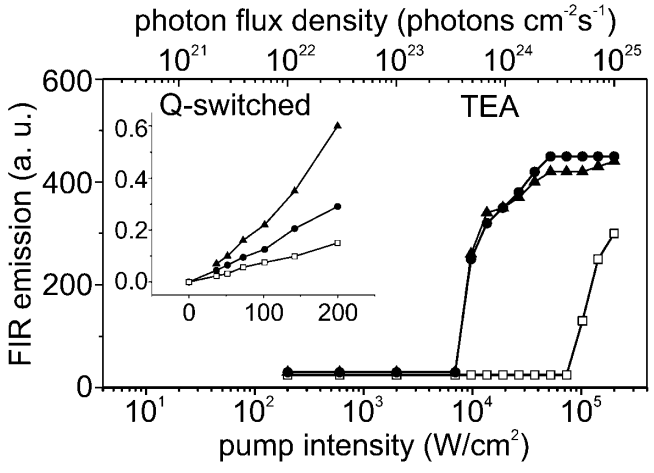

FIG. 2. Dependence of FIR emission from Si:P on the CO_2 laser pump intensity. Squares: sample 2; circles: sample 3; triangles: sample 4. The lower points for TEA laser pumping correspond to the noise level (25 mV).

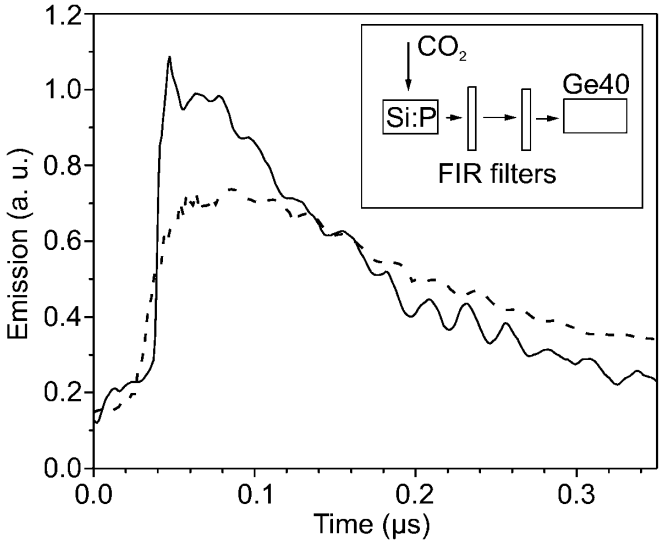

FIG. 3. Traces of the CO_2 laser pump signal (dashed) and the FIR stimulated emission signal (solid) for sample 3. The inset shows the layout for the observation of the stimulated emission.

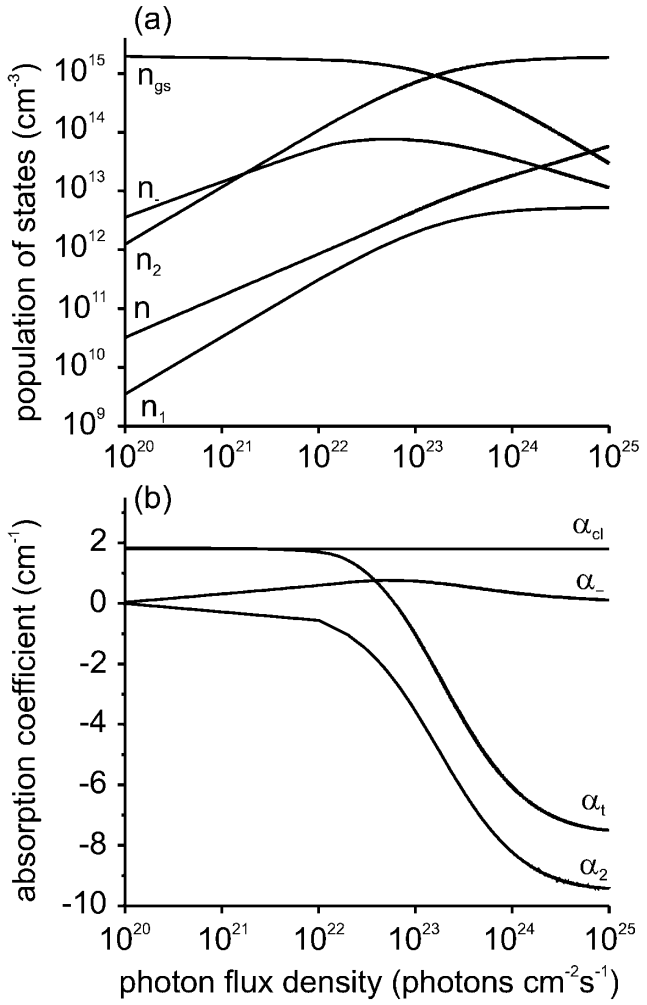

FIG. 4. (a) Calculated population of $1s(A)$ (n_{gs}), $1s(E)$ (n_1), $2p_0$ (n_2), D^- center (n_-) states, and free carrier concentration (n). (b) Calculated absorption and amplification coefficients for the $2p_0 \rightarrow 1s(E)$ transition versus CO_2 laser photon flux density for the parameters of sample 3: α_2: amplification on transitions from the $2p_0$ state, including the absorption by $2p_0 \rightarrow$ continuum transitions; α_-: absorption on D^- centers $\rightarrow$ continuum transitions; α_{cl}: cavity losses; α_t: total amplification.

In conclusion, stimulated emission from the $2p_0 \rightarrow 1s(E)$ transition in silicon doped with phosphor donors under optical excitation with a CO_2 laser has been observed. The population inversion is due to the suppression of acoustical phonon assisted intracenter relaxation between these states. The experimental values of the FIR gain and the threshold CO_2 laser intensity are in good qualitative agreement with the theoretical results. Similar effects are expected in Si:As and Si:Sb as well as in Si:Li. The presented results pave the way for a new class of mid-infrared and far-infrared lasers.

This work was supported by the Deutsche Forschungsgemeinschaft and the Russian Foundation for Basic Research (RFBR) (joint Grants No. 436 RUS 113/206/1 and No. 96-02-00249 G) as well as RFBR Grant No. 99-02-17958 and the grant of the Russian Academy of Sciences for young scientists (No. 33). S. G. Pavlov gratefully acknowledges the Deutscher Akademischer Austauschdienst for financial support.

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