Invited Paper

Terahertz silicon lasers based on intracenter impurity transitions

H.-W. Hübers [1, 2*], S. G. Pavlov [1] , R. Kh. Zhukavin [3], and V. N. Shastin [3]
[1] German Aerospace Center (DLR), Rutherfordstr. 2, 12489 Berlin, Germany
[2] Humboldt-Universität zu Berlin, Newtonstr. 15, 12489 Berlin, Germany
[3] Institute of Physics of Microstructures, Russian Academy of Science, 603950Nizhny Novgorod, Russia
[*2] Email: heinz-wilhelm.huebers@dlr.de

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Abstract: The first silicon laser was reported in the year 2000. It is based on impurity transitions of the hydrogen-like phosphorus donor in monocrystalline silicon. Several lasers based on other group-V donors in silicon have been demonstrated since then. These lasers operate at low lattice temperatures under optical pumping by a mid-infrared laser and emit light at discrete wavelengths in the range from 250 to 50 *μm* (1.2 *THz* to 6.9 *THz)*. Dipole-allowed optical transitions between particular excited states of group-V substitutional donors are utilized for donor-type terahertz (THz) silicon lasers. Population inversion is achieved due to specific electron-phonon interactions inside the impurity atom. This results in long-living and short-living excited states of the donor centers. The frequency of the laser can be tuned by applying an external magnetic field or by applying a compressive force to the laser crystal. Another type of the THz laser utilizes stimulated resonant Raman-type scattering of photons by a Raman-active intracenter electronic transition. By varying the pump laser frequency, the frequency of the Raman intracenter silicon laser can be continuously changed between at least 4.5 *THz* and 6.4 *THz*. Recently lasing from p-type boron-doped silicon has been obtained. In addition, fundamental aspects of the laser process provide new information about the peculiarities of electronic capture by shallow impurity centers in silicon, lifetimes of non-equilibrium carriers in excited impurity states, and electron-phonon interaction.

Keywords: Terahertz, Silicon, Laser, Donor, Impurity, Raman

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1. Introduction

The optically pumped solid state lasers based on optical transitions of atoms incorporated in solid lattices are to date the most powerful, robust and reliable sources. The key characteristic providing such a high laser gain in solids is its extremely long lifetime of the upper laser level, up to a few milliseconds. The prime reason of such long lifetimes is large energy gaps between atomic levels taking part in a laser action. In the terahertz (THz) frequency range, the hundred times smaller energy gaps become in the order of thermal distortions in the lattice. This prevents simple scaling of laser mechanisms successfully realized in the infrared range with solid-state (intracenter transitions) and semiconductor (bandgap transitions) lasers into the THz range. For instance, 1-5 *THz* GaAs-based quantum cascade lasers utilize inversion based on intra conduction band levels with typical lifetimes of a few picoseconds at most [1]. These short lifetimes of electrons in heterostructure semiconductor lasers are caused by delocalization of electron levels, which are electronic subbands with a state continuum at least in one space dimension. The only THz range energy gaps between localized levels available in solids are presented in the bandgap by hydrogen-like impurity centers in semiconductors. Such levels were supposed to have orders of magnitude longer lifetimes than intersubband ones in heterostructures and therefore could potentially serve for solid-state type THz lasing. Low lattice absorption losses in elemental semiconductors, such as silicon and germanium, together with their larger thermal conductivity, were expected to provide minimal optical loss. In this paper we briefly review the last achievements in the development of silicon-based optically pumped intracenter lasers.

2. Methods and results

Silicon is a very attractive candidate for an emitter in the THz range, mostly due to its compatibility with the well-developed CMOS technology. At low lattice temperatures, supposed for the laser operation, the typical optical loss of undoped material does not exceed 0.1/*cm* over the entire THz range [2]. The condition of localization of impurity levels limits doping below $10^{16}/cm^3$, which in turn makes free electron absorption almost negligible. At the beginning of the research on silicon lasers, it was expected that due to the long lifetimes, which at that time were believed to be in the nanosecond scale [3], values for the optical gain of a few cm^{-1} can be achieved under relatively low excitation rates. This would support even quasi-continuous laser operation [4]. Modern silicon technology not only provides a precise doping of natural silicon by certain elements with simultaneous suppression of undesirable electrically active centers but also allows the growth of ideal lattices based on isotopically enriched crystals. As a standard procedure, float-zone crystal growth with simultaneous doping of a crystal from the melt has been used. More sophisticated techniques have been employed for doping of silicon by volatile elements [5] as well as multi-element co-doping [6]. The high refractive index of silicon support high quality optical resonators based on total reflection modes, which is straight forward obtained by optical polishing of crystal facets.

3. Inversion-based silicon lasers under photoionizing pumping

The beauty of a doped semiconductor as a solid state laser is that it can be optically excited with a wide variety of pump sources once the pump photon energy exceeds impurity ionization, in silicon typically shorter than 20-30 μm. The photoionized electrons relax to the bottom of the conduction band. This is followed by an intracenter relaxation down to a long-living impurity state leading to population inversion between this state and the lower states [4]. This simple approach was realized first in 1999 by pumping of silicon doped by phosphorus (Si:P) with a mid-infrared powerful pulsed carbon dioxide laser (CO2) [7]. Surprisingly, pump rates much higher than originally expected (about 10^8/s and higher) had to be used for achieving laser action [8]. Investigations of the reasons causing such high laser pumping intensities led to understanding a number of new features affecting operation of silicon lasers. As one of the inevitable consequences of the photoionizing pumping experiments appears a multi-channel intracenter relaxation of free carriers, which has not been considered before neither theoretically nor experimentally. We have found experimentally that, depending on the donor level structure, different ladder-type relaxation paths can be used by electrons to reach the impurity ground state. This includes paths bypassing the long-living impurity state, which is supposed to act as an upper laser level [9]. This reduces significantly the population inversion in the laser medium even if the lifetime of a particular state is reasonably long, leading to very high optical laser thresholds, up to about 10^{25} $photon/cm^2/s$ in the case of Si:As [10]. Therefore in this material the state with the longest lifetime is not the upper laser level for lasing under photoionization pumping (Fig. 1). Moreover, at low excitation rates relaxation of photoionized electrons in conventional low-compensated silicon crystals suffers from their capture on neutral donor centers, which results in the formation of long-living and broad-band absorbing D^- centers [11]. These factors can be avoided by using intracenter pumping with photon energies below binding energies of particular photons.

4. Inversion-based silicon lasers under intracenter resonant pumping

Since excited impurity states of donors in silicon have a discrete energy structure in the silicon bandgap, optical pumping requires a precise, resonant activation of electrons bound to the donor ground state, $1s(A_1)$. Such a resonant pumping with the required intensity is possible by using a wavelength tunable infrared free electron laser. We have used the FELIX User facility in the Dutch FOM Institute for Molecular Physics in Rijnhuizen (now moved to the Radboud University in Nijmegen) to resonantly pump donor centers in silicon. Starting from the first successful laser action in Si:P in 2001 [12], we have obtained consequently operation of a silicon intracenter laser for all hydrogen-like donors with about twenty different laser schemes (Fig. 1). The lowest laser threshold achieved in the case of resonant pumping in the $2p_0$ state is about 10^{22} *photon/cm^2/s* in the case of Si:P and Si:Sb [13]. Since intracenter pumping avoids ionization of free electrons this eliminates loss processes connected with the electronic capture from the conduction band. As a result, lasing thresholds are significantly reduced. In addition, different states can serve as upper and lower laser levels. Also, larger optical cross-sections for intracenter resonant pumping, up to ~10^{-14} *cm*^{2}, play a positive role for the efficiency of electronic excitation if compared with photoionization, ~2×10^{-16} *cm*^{2} [4].

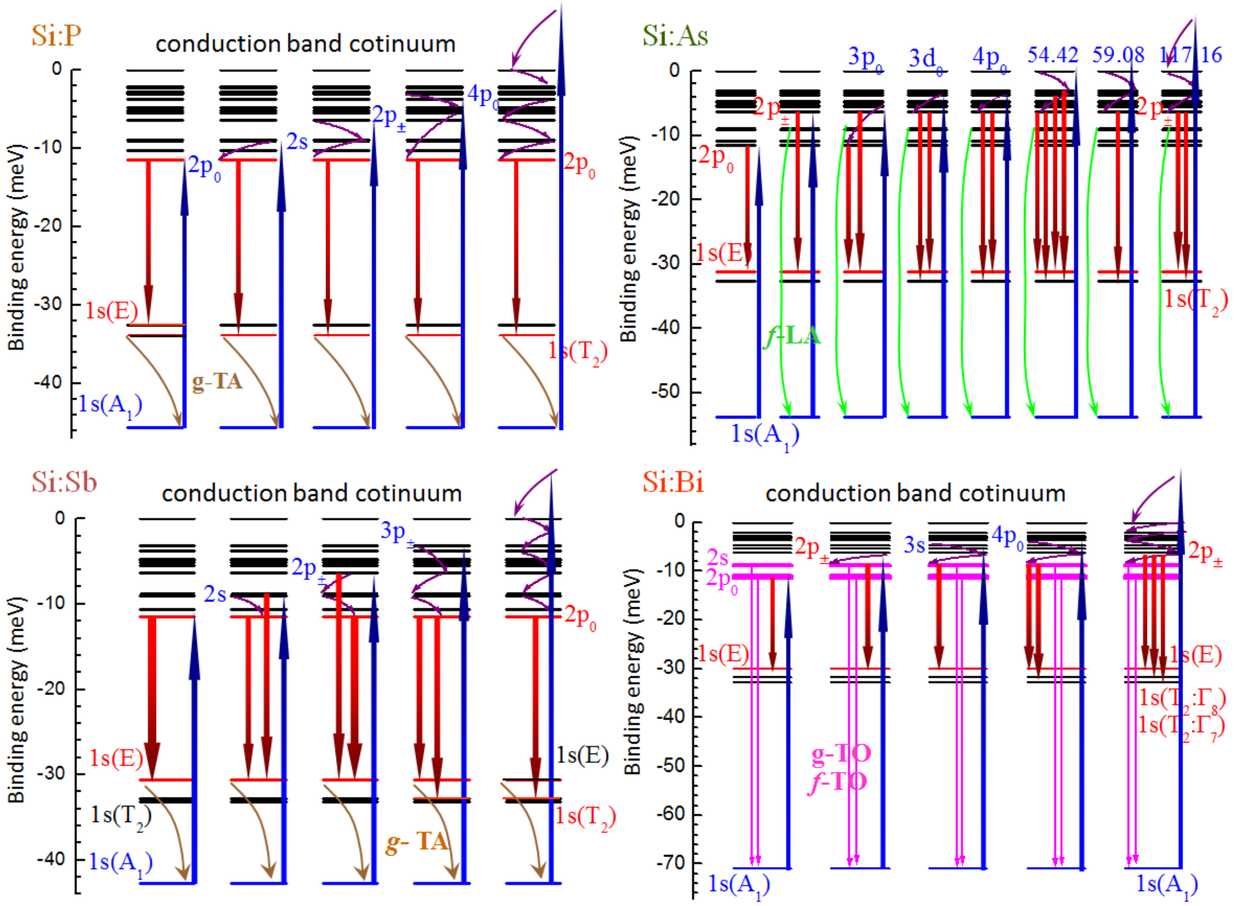

Fig. 1 Variety of the realized silicon laser schemes using photoionizing (pumping in the conduction band continuum) and intracenter (resonant pumping in a localized excited impurity state) pumping for different group-V donors. Straight arrows up are pump emission, straight bold arrows down show stimulated emission from silicon crystals. Other arrows down indicate resonant impurity-phonon interactions influencing on a particular laser scheme.

Note here, that only resonant pumping on the transition with the largest cross section resulted in stimulated emission from boron-doped silicon [14]. So far this is the only p-type silicon THz laser. Figure 2 represents a chart of emission frequencies in the THz region realized by CO_2 laser and free electron laser pumping. Note that the stimulated emission at frequencies in the range from 5 to 7 *THz* has not yet been demonstrated by other solid-state media.

The typical emission intensities for both types of pumping do not exceed a few mW in the pulse peak. The typical duration of the laser pulse is about a microsecond for pumping with a 100 *ns* CO_2 laser and equal to the 5-6 *μs* macropulse duration when pumped by emission of a free electron laser. Continuous generation is achieved despite a 1 ns separation of the 10 *ps* short FELIX micropulses due to the long lifetime of a THz photon in the high-Q cavity of a silicon laser.

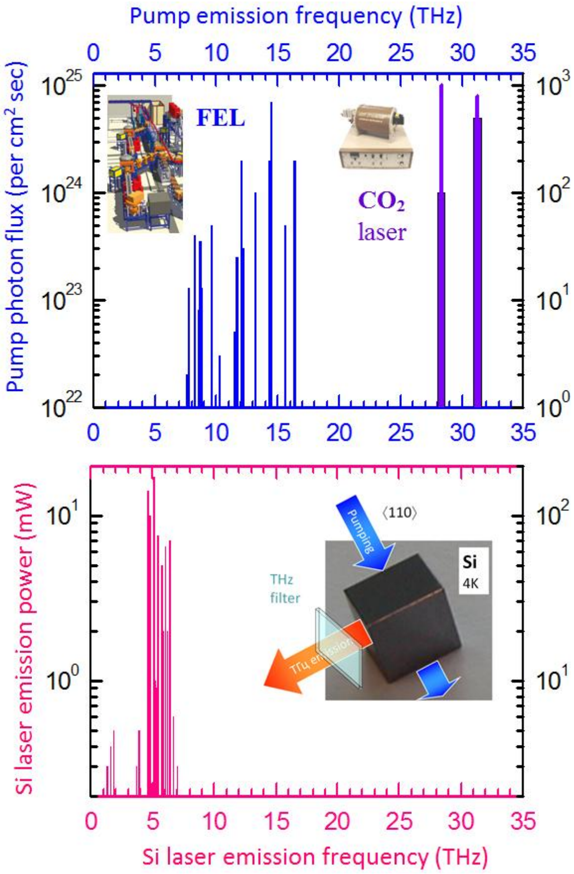

Fig. 2 (upper graph) Pump emission frequencies and thresholds for silicon intracenter lasers under photoionization pumping with a commercial pulsed CO_2 laser and for intracenter pumping with a free electron laser. (lower graph) Laser frequencies and pulse powers of silicon intracenter lasers. The inset shows the geometry of a typical silicon laser pumping scheme.

5. Silicon lasers in the external fields

The properties of THz silicon lasers such as emission frequency and threshold can be changed by external fields or a compressive force which change either the binding energy of the donor or which change donor-phonon interactions. Magnetic fields allow continuous tuning of the binding energies of impurity states due to the Zeeman effect [16]. We have demonstrated a fine, 40-60 *GHz/Tesla*, frequency tunability of the $2p_{\pm}\rightarrow 1s(E)$ and $2p_{\pm}\rightarrow 1s(T_2{:}\Gamma_8)$ laser transitions in Si:Bi [17]. Uniaxial deformations of a silicon crystal can be used for a control of impurity-lattice interactions by tuning in/out of donor-phonon resonances, and by these means, for achievement of longer lifetimes for the upper laser levels or for shortening of the lower laser level. Significant reduction of the lasing threshold have been obtained for all stressed silicon lasers [18, 19], at particular cases uniaxial deformation changes a laser operation scheme [19], for instance in one with a long-living upper laser state in Si:As [20]. Interest has been attracted to the possibility of injection-type excitation of intracenter lasers. However, we have shown experimentally that an electric field destroys population inversion on the donor levels [15] making electrical pumping of silicon impracticable.

6. Inversion-less silicon lasers under intracenter pumping

Beside population inversion based lasing in doped silicon, stimulated emission due to resonant Raman scattering has been observed in n-type silicon [21]. Although Raman emission due to scattering on a zone-centered optical phonon has been demonstrated [22], it was unexpected that Raman-type stimulated emission, vanishing with a wavelength to the fourth power, can be achieved in the THz range. However, resonant scattering at Raman active donor intracenter resonances, $1s(A_1)$-$1s(E)$ transition, made stimulated emission under resonant excitation with a free electron laser feasible for all shallow donors [23]. Lasing at frequencies covering the range of 4.5–6.4 *THz* has been achieved by tuning the pump wavelength between 19 *μm* and 40 *μm*.

7. Conclusion

Over last decade, intracenter silicon lasers have been thoroughly studied in order to show their potential as THz emission sources. It was shown that the factors limiting their efficiency are relatively short lifetimes of the upper laser levels (about 200 *ps* for natural and about 235 *ps* for monoisotopic 28^{th} silicon [24]), the complexity of intracenter relaxation, and inefficient pumping. The largest realized gain values [25] are in the order or even somewhat larger than those achieved with THz quantum cascade lasers, but required large optical pump powers or resonant optical pumping not available in standard laboratories. For inversion-based lasing the best laser performance has been obtained for a neutron doped, compensated silicon crystal under uniaxial stress while for Raman lasing isotopically purified ^{28}Si silicon crystals are the best material.

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