The physics of optically pumped semiconductor bulk lasers for the 5-15 THz frequency range

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The basic physical principles and experimental evidence of terahertz emission from bulk semiconductors doped by shallow impurity centers are reported. The laser mechanism is based on the accumulation of charge carriers optically pumped by a mid-infrared laser, on a particular long-living excited impurity state. Several hundredths of mW of pulsed THz emission output power (optical efficiency around 10^{-6}) has been obtained from elemental (Si, Ge), III-V (InSb) and II-VI (ZnSe) semiconductors at liquid helium temperatures. We expect laser effects for the range 5-15 THz from a number of semiconductors with impurity binding energies in the range 30-130 meV.

1. Bulk semiconductors as new active media for THz light emission

Coherent emission sources for the THz range are required for applications in solid-state spectroscopy, radio astronomy, active imaging as well as environmental monitoring. There is a gross lack of tunable fundamental oscillators in the 5-15 THz range (wavelength range: 20-60 µm). Presently, accessible laboratory optically pumped far-infrared gas lasers only fill the 1-10 THz range in a discrete manner [1] with the most useful operation arising from ca. 20 lines. Narrow band-gap semiconductor lasers reach the mid-infrared as well as the far-infrared region, but operate stably at wavelengths not greater than 3-6 µm [2]. New low-dimensional intersubband quantum “fountain” (17-19 µm) [3] and quantum “cascade” lasers (4-25 µm) [4] are effective only in the mid-infrared region. Fixed lasing frequencies and the necessity for a quasi-optical resonator for operation in the THz range, restricts the use of these lasers. Tunable intersubband hot hole p-Ge lasers [5] cover the wavelength range of 80-250 µm, but with low efficiency (gain ca. 0.2 cm^{-1}).

Recently proposed fundamental oscillators, based on intracentre impurity transitions in bulk semiconductors [6-11], offer significant advantages for applications in the 5-15 THz range. The intracentre optical transitions lie in the THz region and have comparatively high cross sections (up to 10^{-13} cm^2), which implies high gain (1-10 cm^{-1}). Bulk semiconductors are well studied and their manufacture is technologically well developed. The crystals available have geometrical dimensions many times bigger than the expected lasing wavelengths and thus, the crystal itself can serve as a quasi-optical resonator. The lasing output will have a single frequency emission line, which can be tuned by the application of an external magnetic field (frequency fine tuning) or applied stress (broad-band frequency tuning).

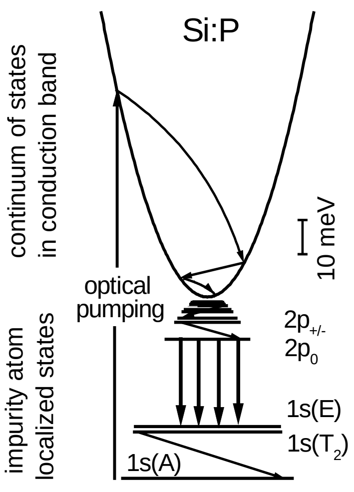
The principle of the laser scheme is common to a number of bulk semiconductors, and is expected to cover the far-infrared range between 18 and 60 µm.

2. Population inversion mechanisms

The overall principle for the formation of a population inversion between excited states of shallow impurity centres in bulk semiconductors is the use of the peculiarities of the relaxation processes, though the system of impurity levels, of the non-equilibrium charge carriers. The non-equilibrium carriers are created by optical excitation of the electrons bound to impurity atoms. At low temperatures (4 K), the dominant process for the relaxation of free charge carriers in semiconductors with relatively low doping concentrations, is non-radiative capture. The carriers, optically excited from the ground state into the continuum of states in the band, first lose their energy in the band by the emission of acoustical or optical phonons. Thereafter, the relaxation of free-charge carriers is via a cascade process. The carrier is first captured in a highly excited state and then gradually relaxes down the ladder of excited states emitting one acoustical phonon during each transition. The characteristic energy step between the neighboring excited impurity states is $\Delta E = (E_i\ m^*\ s^2)^{1/2}$, where m^* is the effective mass, s – velocity of sound and E_i is the ionization energy of i impurity state. The density of states of Coulomb centers drops as $E_i^{-5/2}$. Therefore, the lower excited states are separated by the energy gap exceeding the characteristic step for acoustic phonon assisted relaxation. Such (a) states are $2p_0$ for n-Si, n-Ge and $1P_{3/2}(1\Gamma_8^-)$ for p-Si, p-Ge as well as $2P_{3/2}(1\Gamma_8^-)$ for acceptors in III-V and II-VI crystals - appears as a *long-living* one.

As an exception from this rule, there are materials, where the relaxation between a pair of particular excited states goes via faster (ca. 10^{-12} s) *resonant electron-phonon interaction*, emitting optical phonons. In this case, the upper state of such a pair will be effectively depleted by this resonant relaxation. All excited states in between these interacting states, essentially “switched off” from cascade carrier relaxation, will also have *low populations*.

2.1. Population inversion through suppressed intracentre relaxation



As was described above, deceleration of the cascade relaxation of the non-equilibrium carriers provides the means for a relatively long-living excited state (at 4K). Optical excitation of impurity centres by a laser with photon energy of about or rather higher than the binding energy of the dopant, leads to accumulation of excited carriers in this long-living state. Thus, a population inversion between the long-living state and the lower impurity states is formed. For laser action, corresponding amplification on the intracentre transitions must exceed the internal medium losses (lattice, free electron, impurity-to-band and D^- centres absorptions [9]) as well as the losses on the radiation from the external resonator.

Fig. 1. Scheme of optical (vertical) and non-radiative transitions in Si:P laser. THz lasing (arrows down) occurs from the donor long-living state $2p_0$ to the 1s(T,E) states.

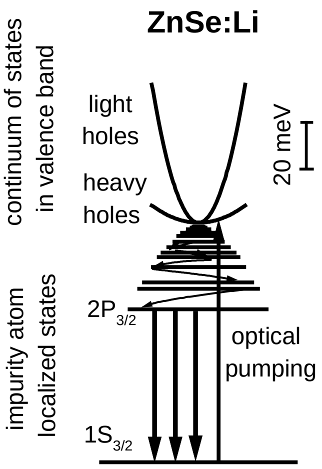


Fig. 2. Scheme of optical (vertical arrows) and non-radiative transitions for ZnSe:Li. THz lasing (arrows down) is expected from the $2P_{3/2} \rightarrow 1S_{3/2}$ optical transitions. $2P_{3/2}$ is acceptor long-living state.

In elemental semiconductors such as n-type Si and Ge, where the ground impurity state (1s) is splitted off by multi-valley interaction, this population inversion mechanism results in 4-level laser scheme (see Fig. 1). This results in lasing at a wavelength of 54 μm from the $2p_0 \rightarrow 1s(T,E)$ transitions in Si doped by phosphor (Si:P), which has recently been observed while pumped by a CO_2 laser [8] and by a free electron laser (IR Facility FELIX, The Netherlands) [11]. Germanium doped by tellurium (Ge:Te) is expected to be an efficient source since Te binding energy (93 meV) fits well to the CO_2 laser quantum energy (main radiation line at 117 meV).

In $A_{III}B_V$ and $A_{II}B_{VI}$ semiconductors as well as Si and Ge, doped by shallow acceptors, this population inversion mechanism might result in a 3-level laser scheme (Fig. 2). The expected laser operation is from optical transitions between the lower excited and ground impurity states.

THz range spontaneous emission has been obtained from several semiconductors, supposedly possessing long-living excited impurity states; Si:B, Ge:Be, Ge:Te, InSb:Zn, InSb:Cu, ZnSe:Li, while irradiated by a CO_2 laser at liquid helium temperatures. Some of the media, such as Ge:Te, InSb:Cu and ZnSe:Li, show a non-linear dependency of the emission with pump power, which suggests THz emission amplification.

2.2. Population inversion through fast intracentre relaxation

A contradictory situation occurs for a few semiconductors, such as Si doped by bismuth and gallium (Si:Bi, Si:Ga) as well as Ge doped by copper (Ge:Cu). For these materials an optical phonon is resonant with the characteristic energy gap between a particular pair of the impurity states. As a result, the upper of the interacting states, for Si:Bi e.g. the $2p_0$ and the 2s states close to it, have a very short lifetimes (ca. 10^{-12} s) and remain depleted unless at high optical excitation rates. Since relaxation due to optical phonons dominates, dumping carriers directly to the ground 1s(A) state, the intermediate 1s(E, T) states populations are negligible (Fig. 3). Hence, a population inversion between the higher excited states and the lower 2s, $2p_0$, 1s(E, T) states is expected [10].

Recently lasing at a wavelength of about 52 μm was obtained from the $2p_{\pm} \rightarrow 1s(E)$ transitions in Si:Bi when pumped by a CO_2 laser [10] and by a FELIX [11] radiation.

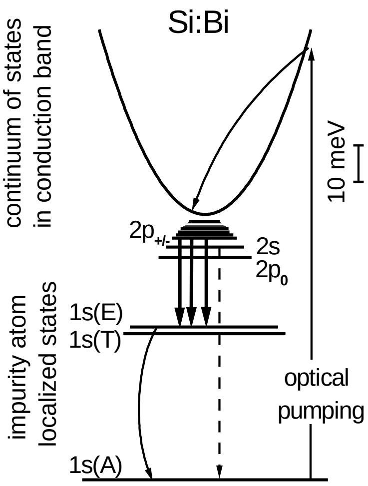


Fig. 3. Scheme of optical (vertical arrows) and non-radiative transitions in Si:Bi laser. THz lasing (arrows down) occurs from the $2p_{\pm} \rightarrow 1s(E)$ transition, while the $2p_0$ and 2s states are depleted by the resonant interaction with ground the state via optical phonon (dashed arrow down). 2s, $2p_0$, 1s(E, T) states have low populations.

3. Conclusions

The research area of interest are bulk semiconductors doped by impurity centers with binding energies of neutral impurity atoms in the range of 30-130 meV. These centers can be effectively excited by radiation of commercial laser sources, such as a TEA or Q-switched CO_2 laser.

To date, evidence of THz range stimulated emission has been obtained from Si:P and Si:Bi crystals, with dominant doping concentrations in the range $(1\text{-}5)\times10^{15}$ cm^{-3} and when optically excited by CO_2 or free electron laser radiation at liquid helium temperatures. Lasing in the THz region was obtained from treated samples (optical quality) with rectangular parallelepiped forms and dimensions of $7\times7\times5$ mm^3. The emission spectra for these media confirm the predicted optical transitions involved, proving the proposed laser schemes for the media.

A number of other bulk materials, doped by shallow level impurities are under investigation at present. Many of them look promising as potential THz light emitters.

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