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To cite this article: A V Andrianov *et al* 2015 *J. Phys.: Conf. Ser.* **647** 012015

Terahertz emission from lithium doped silicon under continuous wave interband optical excitation

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Abstract. We report on experimental observation and study of terahertz emission from lithium doped silicon crystals under continuous wave band-to-band optical excitation. It is shown that radiative transitions of electrons from 2P excited states of lithium donor to the 1S(A1) donor ground state prevail in the emission spectrum. The terahertz emission occurs due to capture of nonequilibrium electrons to charged donors, which in turn are generated in the crystal as a result of impurity assisted electron-hole recombination. Besides the intracentre radiative transitions the terahertz emission spectrum exhibits also features at about 12.7 and 15.27 meV, which could be related to intraexciton transitions and transitions from the continuum to the free exciton ground state.

1. Introduction

During last two decades, terahertz (THz) sources of electromagnetic radiation with frequencies between 0.1 and 10 THz have attracted attention for many applications such as remote sensing, bio-chemical detection, communication, and medical diagnostics. One of the possible schemes of a relatively simple THz emitter is based on optical transitions between energy levels of shallow impurities in semiconductors. The intracenter THz radiative transitions appear at the energy relaxation of nonequilibrium carriers created in the allowed band, e.g. at the impact ionization of impurities in the electric field [1-4] or at the photoionization of impurities by infrared laser radiation [5]. THz lasing on intacenter optical transitions in germanium and silicon was demonstrated in [2, 5]. It has recently been shown that the impurity related THz radiation can also appear under interband optical excitation of semiconductors doped with shallow centres [6]. Such a THz photoluminescence (PL) was detected in a number of materials [6-8]. The THz PL is due to features of electron-hole recombination involving impurity states. Such a recombination results in the formation of charged impurity centres and free carriers in the allowed band. The subsequent capture of free carriers to charged centres is accompanied by THz radiation [7, 8] in similar way as it occurs under electrical breakdown of impurity centres [1-4]. The THz PL in semiconductors can be quite intense and it can find applications in THz technologies. For example, it might be more convenient to use the band-to-band optical

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pumping to excite intracenter radiative transitions when the impurity breakdown requires high electric fields (e.g. in Si, where impurity breakdown threshold field exceeds 1000 V/cm [3]).

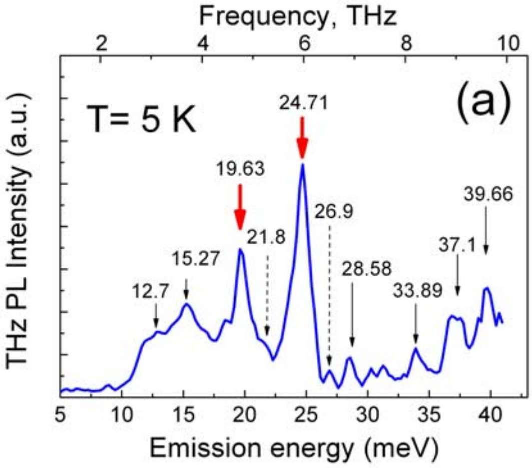
THz emitters based on impurity optical transitions in silicon are of interest because they can be directly integrated with silicon electronics. Lithium in silicon demonstrates quite a few unusual properties comparing with other donor impurities. One can mention an ability of lithium to form complexes with others impurities, which exist in the crystal. The ionization energy of Li donor in Si is substantially lower than that for others donors, for example, formed by V group elements. The electronic structure of the Li donor ground state is also unusual. There is so called "inverted" in comparison with V group donors arrangement of the sublevels of the Li donor ground state [9]. Lithium in Si is one of possible candidates for a Qbit in a quantum computer with the information readout based on intracenter optical transitions [10]. In view of said above, it is of interest to investigate intracenter THz luminescence associated with Li donors in Si.
This paper reports the detection and study of THz PL from Si:Li crystals.

2. Experimental details

2.1. Samples under the test
The experiments were curried out on silicon single crystals doped with lithium to a level of 10^{16} cm^{-3}. The crystals were doped during the growth from melt by the method of growth from a pedestal (a variant of float-zoning technique). According to measurements of optical absorption in the middle infrared range, the concentration of oxygen was below 10^{16} cm^{-3}. The samples for measurements were prepared in the form of chemically-mechanically polished 1.5-mm-thick 5x7-mm plates.

2.2. THz measurements
The Si:Li samples were placed in a helium optical cryostat with a controlled temperature, which was optimized for the THz spectral domain. A CW semiconductor laser emitting at λ=660 nm with maximal output power of about 45 mW was used as a source of the interband photoexcitation. The intensity of photoexcitation did not exceed 1 W/cm^2. The main measurements of THz PL were performed in "transmission" geometry. Some control experiments were also performed in "backscattering" geometry. The results of these measurements were almost the same. The spectral measurements were done with using a step-scan Fourier spectrometer for spectral range of 5-350 cm^{-1}, described elsewhere [4]. The spectral resolution in most cases was 5 cm^{-1} (0.62 meV). A THz radiation signal was measured with liquid helium cooled Si bolometer by synchronous detection method at the laser-radiation modulation frequency by a mechanical interrupter (80 Hz).

3. Results and discussion
The characteristic spectrum of the THz PL from Si:Li crystal observed at liquid helium temperatures is shown on the figure 1a. It is seen that the emission lines with maxima at 24.71 meV (5.98 THz) and 19.63 meV (4.75 THz) dominate in the spectrum. The difference between the energies of these lines is 5.08 meV. This value is very close to well-known energy difference of 5.11 meV between the $2P_{\pm}$ and $2P_0$ levels of the donor impurity in silicon, which was thoroughly calculated and confirmed experimentally for various donor centres in Si (see Ref. [9]). Besides that the position of the 24.71-meV emission line is in good agreement with the position of the line of the transition between the $2P_{\pm}$ excited state and $1S(A_1)$ sublevel of the ground state of Li donor in silicon, which was observed in far infrared absorption experiments [9]. It allows us to attribute the intense emission lines at 24.71 and 19.63 meV to intracenter transitions $2P_{\pm}\rightarrow 1S(A_1)$ and $2P_0 \rightarrow 1S(A_1)$ in Li donor, respectively (see figure 1b).
The character of the THz PL spectrum indicates that the lines of radiative transitions from 2P states to the lowest sublevel $1S(E+T_2)$ of the ground state of Li donor are significantly suppressed as compared to the lines of transitions to the $1S(A_1)$ sublevel.



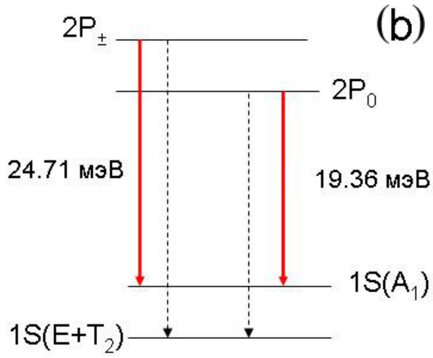


Figure 1. a) Typical THz PL spectrum at I_{exc}~ 1W/cm^2. Arrows point to the positions of THz emission lines. The labels show the energies in "meV". b) Scheme of main intracenter radiative transitions.

Weak line at 26.9 meV and a feature at the energy of ~21.8 meV, which looks as a high-energy shoulder of the line of the $2P_0 \rightarrow 1S(A_1)$ transition, marked by dashed arrows in the figure 1, are most likely related to transition to the $1S(E+T_2)$ ground state of Li donor from the $2P_{\pm}$ and $2P_0$ excited states, respectively. Strong suppression of the THz emission associated with transitions to the $1S(E+T_2)$ state as compared to emission caused by transition to $1S(A_1)$ state can be attributed to the effect of reabsorption of the radiation. Indeed, due to the total internal reflection only a small fraction of the radiation is directly emitted from the excitation region. The main fraction of radiation, multiply reflected from the surface of the crystal, travels inside it and undergoes absorption. At helium temperatures, the lowest sublevel $1S(E+T_2)$ of the donor ground state is populated much higher than the sublevel $1S(A_1)$. Therefore, the absorption of THz radiation on the lines corresponding to the $2P \rightarrow 1S(E+T_2)$ transition is much higher than on the lines corresponding to the $2P \rightarrow 1S(A_1)$ transitions. We have to note that, in the experiments on THz electroluminescence (EL) under impurity breakdown in Si:P [3], similar regularity was observed. The authors have observed significant (almost complete) suppression of the emission associated with transitions to the lowest sublevel of the donor ground state as compared to the emission associated with transitions to higher lying sublevels of the donor ground state. But this feature was not explained in [3]. We believe that this feature of the THz EL spectra in Si:P can also be explained by the THz radiation reabsorption effect.

Weak emission lines in the THz PL spectrum of silicon with lithium in the energy range from 27.5 to 40 meV (see figure 1a) can be attributed to intracenter optical transitions in donors caused by Li-O complexes. The lines with maxima at 28.58 and 33.89 meV are due to optical transitions to the ground state $1S(A_1)$ of the Li-O donor from the $2P_0$ and2 $P_{\pm}$ excited states, respectively. Wider line with the maximum at about 37.1 meV can be attributed to the superposition of transitions from $4P_{\pm}$ and $3P_{\pm}$ states to the $1S(A_1)$ state, and the 39.66 meV line is possibly due to transitions of electrons from the conduction band to the $1S(A_1)$ state. Such an interpretation is in good agreement with the data on the spectrum of energy levels of the Li-O donor in silicon [9].

The THz PL spectrum (figure 1a) also exhibits relatively intense features at energies of 12.7 and 15.27 meV. It is hard to assign these features to intracenter transitions in the lithium donors. On the other hand, it is known that in the energy range of 10-14 meV the absorption associated with $1S \rightarrow 2P$ transitions in free excitons in Si is observed [11]. Furthermore, the binding energy of free exciton in Si is known to be about 15 meV [12]. The capture of free carriers to excitons can be accompanied by THz radiation similar to radiation at the capture of carriers into ionized impurity centres. Therefore, we tentatively attribute the 12.7- and 15.27-meV THz emission lines to intraexcitonic radiative transitions and transitions from continuum to the free exciton ground state.

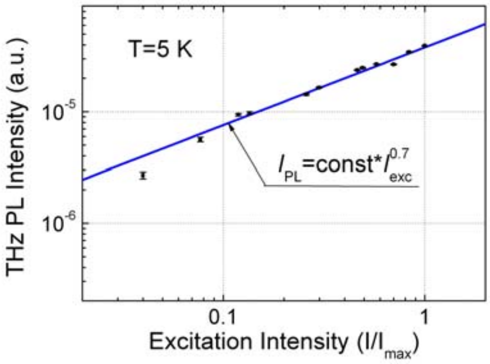


Figure 2. The total PL intensity versus the excitation intensity. I_{max}=1 W/cm^2.

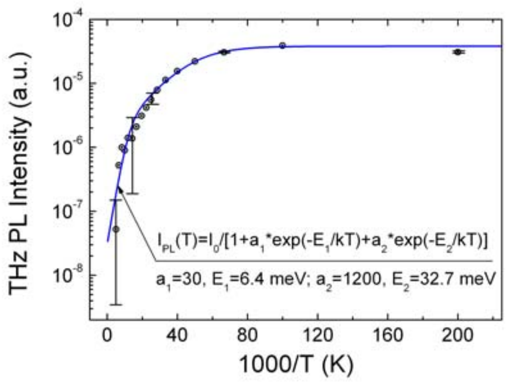


Figure 3. Temperature dependence of the THz PL. The solid curve is a result of fitting of the data.

It was revealed that the THz PL intensity is sublinear versus the excitation intensity (see figure 2). At the excitation intensity above 0.1 W/cm2 this dependence can be well approximated by the power law of I_{PL}~$I_{exc}^{0.7}$. This fact is important and it indicates that the main THz radiation is formed as a result of recombination of nonequilibrium holes with electrons on neutral donors. An analysis, similar to that is given in [8], shows that a sublinear dependence of the THz PL intensity on the excitation intensity should be expected in the case of this mechanism of THz emission.

The THz PL decreases with increase in temperature. Figure 3 shows the temperature dependence of the total intensity of the THz PL in Arrhenius coordinates. As is seen the data can be well approximated by usual activation equation with two temperature quenching energies: E_1=6.4 meV and E_2=32.7 meV. The energy of 32.7 meV is consistent with the binding energy of Li donor in Si [9]. Therefore, the main temperature quenching of the THz PL can be attributed to the temperature dissociation of neutral Li donors. The energy of 6.4 meV is in agreement with binding energy of $2P_{\pm}$ excited state [9]. Most likely this quenching energy reflects a thermal release of electrons from the 2P donor states since transitions from 2P states give significant contribution to the observed THz PL.

4. Conclusion

THz luminescence from Si:Li crystals has been detected and studied under interband optical excitation. It is shown that the main THz PL is caused by intracenter transitions from 2P excited states to the $1S(A_1)$ sublevel of the ground state of the Li donor. The experimental data indicate that the main radiation is formed as a result of recombination of nonequilibrium holes with electrons on neutral donors. The THz PL spectrum also exhibits lines, which are possibly due to intraexcitonic radiative transitions and transitions from continuum to the ground state of excitons.

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