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Stimulated Terahertz Emission of Bismuth Donors in Uniaxially Strained Silicon under Optical Intracenter Excitation

R. Kh. Zhukavin[a,]*, **S. G. Pavlov**[b], **A. Pohl**[c], **N. V. Abrosimov**[d], **H. Riemann**[d], **B. Redlich**[e], **H.-W. Hübers**[b,c], and **V. N. Shastin**[a]

[a] *Institute for Physics of Microstructures, Russian Academy of Sciences, Nizhny Novgorod, 607680 Russia*
[b] *Institute of Optical Sensor Systems, German Aerospace Center (DLR), Berlin, 12489 Germany*
[c] *Department of Physics, Humboldt-Universität zu Berlin, Berlin, 12489 Germany*
[d] *Leibniz-Insitut für Kristallzüchtung (IKZ), Berlin, 12489 Germany*
[e] *Radboud University Nijmegen, Institute for Molecules and Materials, FELIX Laboratory,*
6525 ED Nijmegen, The Netherlands
**e-mail: zhur@ipmras.ru*

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Abstract—The results of the experimental observation of stimulated terahertz emission under optical intracenter excitation of uniaxially strained bismuth-doped silicon are presented. Pumping in the presented experiment is performed using a FELIX free-electron laser. It is shown that uniaxial strain of the silicon crystal leads to a significant change in the stimulated emission spectrum of the impurity.

Keywords: silicon, bismuth, uniaxial pressure, intracenter optical excitation, emission-frequency tuning

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1. INTRODUCTION

In recent years, considerable progress has been attained in the development of semiconductor emission sources of the terahertz frequency range [1]. Nevertheless, the necessity of the correspondence to the requirements of a specialization dictates the importance of certain characteristics of an emission source. One such characteristic is emission line tuning, which can be performed by a method depending on the physical properties and characteristics of the active medium. In this work, the influence of uniaxial deformation on the simulated terahertz emission in donor-doped silicon under intracenter excitation is investigated. Bismuth (Bi) which has an ionization energy of about 71 meV and an active Raman transition with an energy of about 40 meV was selected as the active impurity. The Stokes shift for shallow donors in silicon is determined by the energy difference between $1s(A_1)$ and $1s(E)$ states [2]. It was previously shown that the optical excitation of Group-V donors in silicon at low temperatures (below 30 K) can, depending on the excitation quantum, lead to the appearance of stimulated emission according to two mechanisms, namely, population inversion and stimulated Raman scattering (SRS) [3]. The lasing transitions of the inversion mechanism in the case of the photoionization of Bi are $2p_{\pm} \rightarrow 1s(T_2)$ and $2p_{\pm} \rightarrow 1s(E)$. Population inversion is formed due to the specific relaxation character of excited states of bismuth, at which $1s(T_2)$ and $1s(E)$ states turn out to be relatively unoccupied due to rapid “depopulation” of the $2p_0$ state as a result of the interaction with intervalley optical phonons *f*-TO. Stimulated Raman scattering is observed in the case of intracenter excitation at the $1s(A_1)$–$1s(E)$ transition in a range of pump quanta limited by the energy of the $1s(A_1)$–$2p_0$ transition from below and the $1s(A_1)$–$3p_0$ transition from above [2]. Herewith, both the inversion and Raman mechanisms can be implemented in the case of resonant pumping and the competition of these mechanisms can be observed [3]. The Raman activity at the $1s(A_1)$–$1s(E)$ transition is caused by the wave functions of $1s$ states possessing an evenness relative to the replacement of k_i by $-k_i$, where i is the crystallographic direction along the valley axis of the conduction band of silicon. On the other hand, the $1s(A_1)$–$1s(T_2)$ transition in silicon donors is not Raman active. Another necessary SRS condition is the occurrence of a relatively rapid relaxation channel $1s(E)$, which is provided by the emission of intervalley LA-*f* phonons in this system. Uniaxial deformation of the crystal should lead to splitting of the $1s(E)$ state into two components $1s(B_1)$ and $1s(A_1^{\text{upper}})$ and, consequently, to the presence of two active Raman transitions (in a definite range of deformations for a specific direction) (Fig. 1). In addition, it is known that the

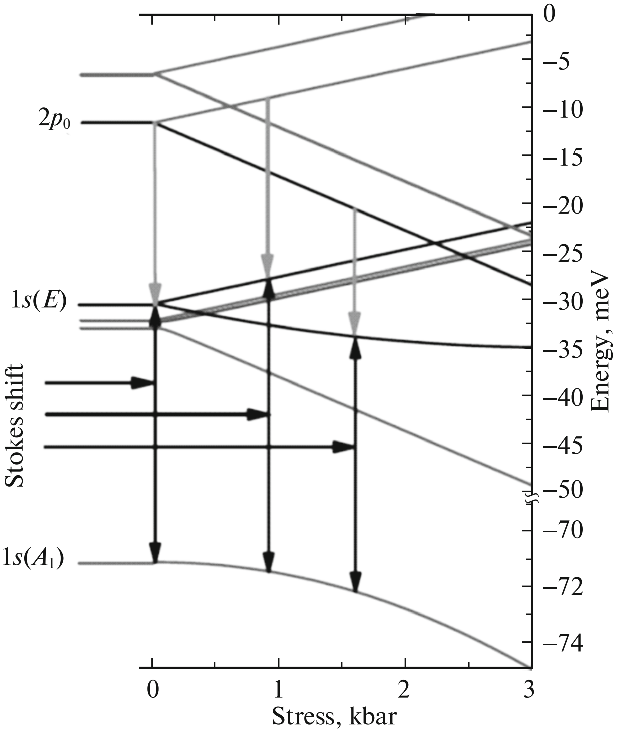

Fig. 1. Splitting of the energy levels of the bismuth donor in silicon under the effect of uniaxial deformation along the [001] crystallographic direction. Arrows directed downward show the emission quantum in the case of the resonant pumping of donor states. Horizontal arrows point to a Stokes shift, whose variation is caused by the splitting and mutual shift of components of the $1s(E)$ and $1s(A_1)$ state. The position of levels is calculated according to [5]. The zero energy value corresponds to the so-called center of "masses" of valleys and coincides with the conduction-band bottom in the absence of pressure.

conditions determining the population inversion in Si:Bi also vary under deformation [4].

2. EXPERIMENTAL

The doping of initial silicon grown by float zone technique was performed by the pedestal-growth method [6]. The bismuth concentration in the crystal was $N_D \sim 3 \times 10^{15}$ cm^{-3}. The sample was cut in the form of a rectangular parallelepiped with sizes of 7 × 5 × 2 mm^3 and a long side along the [001] direction and "optical" quality of face polishing for the formation of a high-Q resonator. The Si:Bi samples were characterized by measuring the impurity absorption at low temperatures (~5 K) using a Fourier spectrometer (Bruker Vertex 80v) with a spectral resolution of 0.1 cm^{-1}. An experiment on the observation of the laser effect was performed using the FELIX user station of a free-electron laser (FEL) (Radboud University, Nijmegen, Netherlands). The pump pulses were macropulses with a duration of 6 μs successive with a frequency of 10 Hz. Each macropulse consists of micropulses with a duration of ~10 ps and a power up to 10 MW separated by a time interval of 1 ns. The pulsed character of pumping emission assumes that the photon lifetime in the resonator is no smaller than the repetition period of excitation micropulses by an order of magnitude. This condition is fulfilled rather well for the used samples; previous experiments showed that the photon lifetime in the resonator is ~10 ns [7]. A special insert containing the sample under study and permitting the application of pressure [8] was immersed into a transport Dewar helium vessel. The excitation spectrum and emission spectrum were measured at various applied pressures. The emission spectrum was measured using a Fourier spectrometer with a spectral resolution of up to 0.5 cm^{-1} and coupled with a Ge:Ga photodetector (the sensitivity band taking into account crystalline quarz as a filter was 40–120 μm). The excitation spectrum (signal of the germanium detector depending on the excitation quantum) was recorded using the software of the FEL FELIX user station.

3. RESULTS AND DISCUSSION

Our experiments allowed us to reveal THz emission from Si:Bi, which has a threshold dependence on the pumping intensity, confirming its stimulated character. The analysis of our experimental data allowed us to reveal contributions that have different dependences on the pressure applied along the [001] direction in the spectrum of the stimulated emission of bismuth in silicon. The excitation spectrum in the absence of deformation corresponds to the transition from the ground state into the $2p_\pm$ state, while the emission spectrum corresponds to the $2p_\pm$–$1s(E)$ transition.

The absence of a broader excitation spectrum in our experiments is caused by a relatively low excitation power and path losses. Generally speaking, the observation of stimulated Raman scattering (SRS) is possible in such a system in the range of pump quanta of 59–65 meV (19–21 μm). Uniaxial deformation leads to splitting of the $2p_\pm$ line into two components, which is expressed in the appearance of similar components in the excitation spectrum at low pressures (Fig. 2). Resonances corresponding to the transition into components of the $2p_0$ state are found in the excitation spectrum at a pressure of >200 bar, while $2p_0$–$1s(A_1^{\text{upper}})$ intracenter transitions are found in the output emission spectrum.

We found the emission frequency tuning for the $2p_0$–(A_1^{upper}) transition with an increase in pressure, which reflects a variation in the energies of the corresponding active transition. On the one hand, the energy of the $2p_0$–(A_1^{upper}) transition in deformed silicon can theoretically vary in rather broad limits (Fig. 1), but the magnitude of the matrix transition element, which should decrease with an increase in pressure, should also be taken into account. The latter is caused by a decrease in the contribution of lower valleys into the wave function of the $1s(A_1^{\text{upper}})$ state; however, in view of the considerable magnitude of the

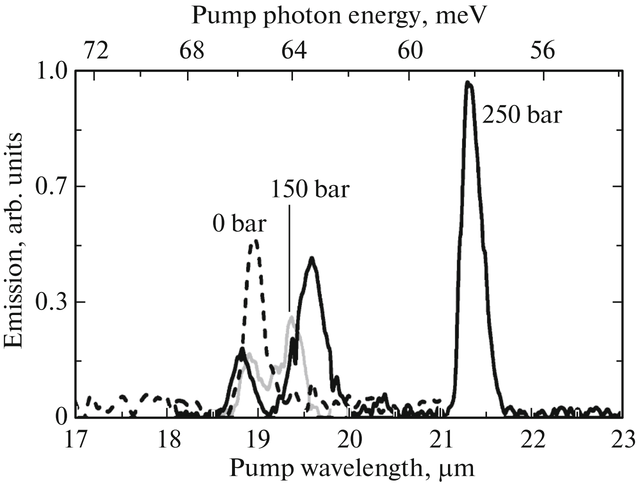

Fig. 2. Dependence of the output intensity of Si:Bi on the excitation wavelength for several values of the pressure along the [001] direction. The intensity of the pumping emission was ~1 MW/cm^2 in a micropulse at the input of the cryogenic insert. The sample temperature is 4.2 K.

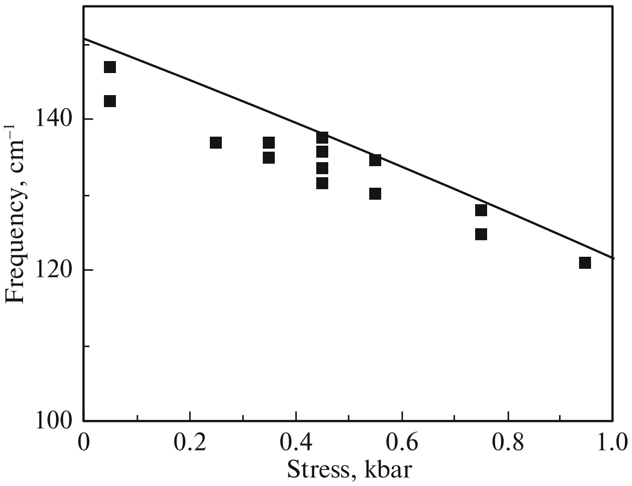

Fig. 3. Dependence of the output emission frequency at the transition between the lower component $2p_0$ and $1s(A_1^{\text{upper}})$ depending on the pressure along the [001] crystal axis under the resonant excitation of $2p_0$. Experimental data are shown by points, and the calculated dependence of the energy of the $2p_0$–$1s(A_1^{\text{upper}})$ transition is shown by a solid line.

"chemical" shift in bismuth, this circumstance can be neglected for pressures used experimentally. In our experiments, the minimum of the observed frequencies of ~120 cm^{-1} is determined by the red boundary of the Ge:Ga-based photodetector and was attained for the $2p_0$–$1s(A_1^{\text{upper}})$ transition (Fig. 3). It should be noted that transitions into the lower component $1s(T_2)$ in our experiments are observed not at all pressures. This circumstance is associated with broadening the levels because of nonuniformity of the applied stress and resonances with intervalley phonons. Both these circumstances should lead to stronger suppression of the inversion mechanism. Indeed, the experiments with donor photoionization [7] showed that lasing is lacking in the range of 1–2 kbar for the uniaxial-deformation direction along [001] [9]. The authors assume that in the case of $2p_0$–$1s(A_1^{\text{upper}})$ transitions, the responsible mechanism is the stimulated Raman scattering of light in view of the stability with respect to the nonuniform broadening of transition lines.

4. CONCLUSIONS

In this work, the influence of uniaxial pressure on the terahertz laser generation in bismuth-doped silicon under the optical intracenter excitation is investigated. It is shown that uniaxial pressure leads to a variation in the characteristics of the stimulated emission. Tuning of the emission line for the $2p_0$–$1s(A_1^{\text{upper}})$ transition in the range of 120–150 cm^{-1} is found.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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