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First terahertz-range experiments on pump – probe setup at Novosibirsk free electron laser

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Abstract

A single-color pump-probe system has been commissioned at the Novosibirsk free electron laser. The laser emits a tunable monochromatic terahertz radiation. To prove the proper system operation, we investigated the time-resolved absorption of a sample of n-type germanium doped with antimony, which was previously investigated at the FELBE facility, in the temperature range from 5 to 40 K. The measured relaxation time amounted to about 1.7 ns, which agreed with the results obtained at the FELBE. The results of pump-probe measurements of non-equilibrium dynamics of hot electrons in the germanium crystal at cryogenic temperatures are presented for wavelengths of 105, 141 and 150 μm.

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1. Introduction

Study of fast processes with high temporal resolution is one of the important problems in the modern science, and the pump-probe technique is a powerful method for solving this task. This technique can be implemented in different variants: the pump pulse and the probe pulse have different wavelengths or, alternatively, the same pulse split with a beam splitter is used for both the pump and probe radiations. Most pump-probe experiments in the terahertz spectral range were carried out using the time-domain technique, in which the pump radiation has an extremely wide spectral width. As an example we can mention the paper by Hoffmann et al. (2009), in which a THz pump – THz probe time-domain experiment is described. In certain cases, however, when excitation of selected levels is required, it is necessary to apply systems with monochromatic pump radiation. Since sources in such experiments are required to have a high pulsed power and a narrow linewidth, as well as radiation wavelength tuning within a wide spectral range, free electron lasers are devices meeting these criteria well.

The pump-probe technique is widely used for study of semiconductors. In paper Deßmann et al. (2015) TEMP per by the recombination time of extrinsic Ge photodetectors was measured in a single-color pump-probe system, which operates at the free electron laser FELBE (Helmholtz Zentrum Dresden-Rossendorf). Now, a similar THz pump – THz probe system is being commissioned at the Novosibirsk free electron laser (NovoFEL) (Kulipanov et al., 2015) at Budker Institute of Nuclear Physics SB RAS.

NovoFEL is a user facility consisting of three laser systems emitting monochromatic high-power radiation in spectral ranges from 5 to 240 μm. The first THz laser system has been in operation since 2003. It emits radiation as a continuous stream of 100-ps pulses with a repetition rate of 5.6 MHz in the spectral range from 90 μm to 240 μm. The average power of the laser beam reaches 500 W at a repetition rate of 11.5 MHz. In a routine regime, the average power of radiation at the user stations is 50-100 W at $\lambda = 130$ μm and $f = 5.6$ MHz.

2. Experimental setup

The equipment of Siberian Center for Synchrotron and Terahertz Radiation was used in the experiments performed. A schematic representation of the pump-probe system is shown in Fig. 1.

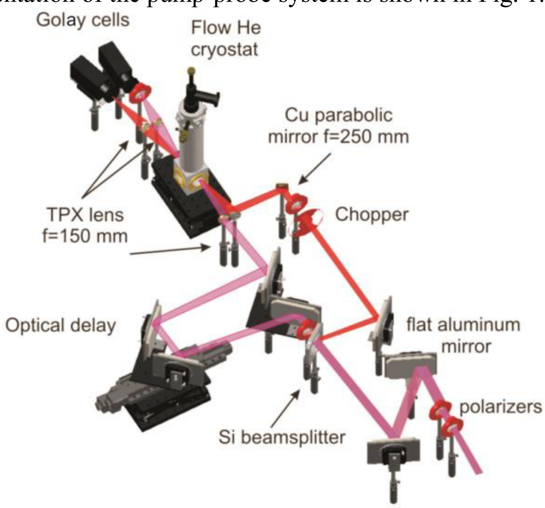

Fig. 1. Schematic presentation of single-color pump-probe setup at NovoFEL facility. Red line: pump beam, purple line: probe beam.

The input NovoFEL radiation was attenuated and polarized with two polarizers. Then a silicon beam splitter divided the radiation into the pump and probe beams with the intensity ratio of 100/1. Additional polarization chengers and attenuators, which are not shown in the figure, enabled independent variation of the intensities of the beams.

Once the probe beam passed the optical delay line, in which a corner reflector was placed on a 300-mm long motorized translation stage, a lens with a focal length of 100 mm focused the beam onto the sample into a spot of 1 mm in diameter. The sample was mounted in a liquid-He-flow Janis-100 cryostat with TPX windows. The temperature of the sample could be set to a value from 5 K to 50 K. A gold-coated parabolic copper mirror with a focal distance of 250 mm focused the pump radiation on the sample surface into a spot of 2.5 mm in diameter. These beams intersected at an angle of 15 degrees. A 320×240 microbolometer array (Dem'yanenko et al., 2008) imaged the cross-sections of the beams, which allowed us to control the positions of the beams on the sample.

Two imaging systems collected the radiation that passed the sample and focused it into two Golay cells. Such configuration of the detector system let us avoid background noise and collect both pump and probe radiations that passed the sample into the input windows of the detectors. To minimize the contribution of the pump radiation portion leaking into the probe beam line because of scattering and reflections, we used orthogonally polarized the pump and probe beams and set an additional blocking polarizer in front of the Golay cell in the probe beam line. The pump beam was chopped with a frequency of 15 Hz, and an SR-830 lock-in amplifier detected the probe beam at the pump-beam modulation frequency, used as a reference one.

3. Results

Czochralski-grown germanium crystal doped with antimony up to 10^{15} cm^{-3} of impurity concentration and negligible residual compensation ~ 10^{12} cm^{-3} was investigated. This Ge:Sb sample was previously studied using the FELBE pump-probe setup by Deßmann et al. (2014). The time of decay of the pump-probe signal at donor ionizing photon energy of 11.8 meV (105 μm) was measured at temperatures from 5 to 40 K for different radiation intensities when photon flux density changed from 10^{26} cm^{-2} s^{-1} to 4×10^{24} cm^{-2} s^{-1}. At a temperature of about 5 K the time constant of the process decreases gradually from 1.7 ns to 1ns with the increasing of the pump power. The dynamics of absorption of the Ge:Sb sample at the same photon energy recorded using the pump-probe system at the NovoFEL is presented in Fig. 2. One exponential function can fit the pump-probe signal decay. The relaxation time for the Ge:Sb sample appeared to be about 1.7 ns. Due to the larger NovoFEL pulse duration, the front slope of the pump-probe signal is less steep as compared with the signal obtained at the FELBE. The agreement between FELBE and NovoFEL results proves the adequacy of selected technique.

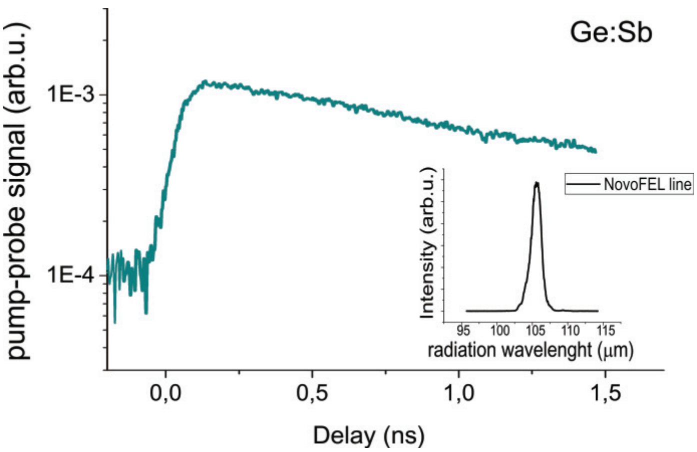

Fig. 2. Pump-probe signal vs. delay time for Ge:Sb sample at 105 μm wavelength, recorded at NovoFEL. Sample temperature: 5 K. Inset: NovoFEL emission line.

In the course of further experiments, we have studied the pump-probe temperature depended signals for two bound-bound transitions for different temperatures at wavelengths of 150 µm and 141 µm, which corresponded to the excitation of the $2p_{\pm}$ (Fig. 3) and $3p_{\pm}$ (Fig. 4) states of antimony in germanium, respectively. It has to be emphasized here that the transitions from the triplet state 1s (T_2) predominate over the transitions from the ground 1s (A_1) singlet state since the valley-orbit splitting is very small ~0.316 meV (Baker and Fisher, 1996). Comparison of the results of the measurements of the decay times for excitation of the localized states suggests relaxation times of ~0.8 ns for the $3p_{\pm}$ state and of τ~1.2 ps for the $2p_{\pm}$ state, *i. e.* the excitation of the higher-lying $3p_{\pm}$ state provides a shorter response time.

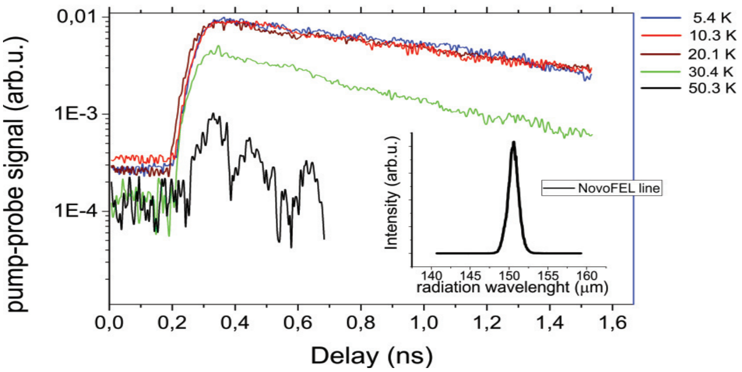

Fig. 3. Pump probe signal vs. delay time for Ge:Sb sample at 150 µm wavelength at different sample temperatures.

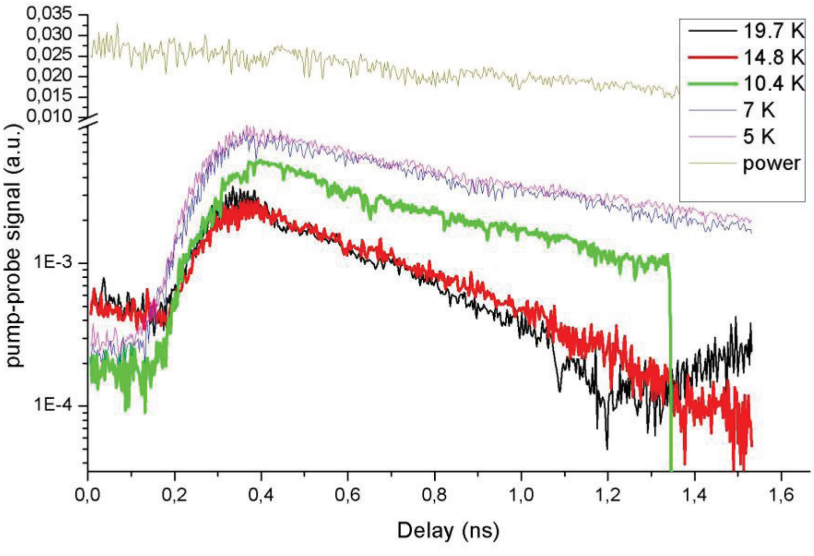

Fig. 4. Pump probe signals vs. delay time for the Ge:Sb sample at 141 µm wavelength at different sample temperatures; the upper signal demonstrates variation of input power during one of the experiments.

4. Acknowledgments

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