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Pump-probe investigations of THz transitions in Si/Si:Er^{3+} nanolayers

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Abstract

A possibility to realize optical transitions within the $^4I_{15/2}$ ground state of Er^{3+} ion in Si, between levels split by crystal-field, has been investigated by pump-probe technique. The study has been conducted in the THz range on a sublimation MBE-grown Si/Si:Er multinanolayer structure, which allows to take advantage of the preferential formation of a single type of Er-related centers. We present preliminary results, which show absorption band around the wavelength of 43 μm. A resonant transition at this wavelength is predicted for this material from high-resolution photoluminescence measurements. The experimentally observed absorption decay time is most likely due to non-radiative recombination by phonon emission.

Keywords: Photoluminescence; Semiconductors; Rare earths

1. Introduction

The THz frequency range remains relatively unexplored. This is because it is positioned at the border between electronically and optically attainable domains, which makes it difficult both for generation and for detection. Such a situation is regretful, since THz radiation finds multiple uses in fundamental research as well as in many practical applications. In fundamental research, THz spectroscopy is a dedicated tool for, e.g., investigations of conductivity mechanisms in solids, since scattering times impeding carriers flow in these materials is typically in the femtosecond range. It is believed that studies of THz interactions will provide new insights into a whole range of materials, including not only solids and liquids, but also polymers and biological tissues. On the application side, in communications, the THz range offers greater bandwidth and reduced antenna sizes. Other commercial uses comprise thermal imaging and remote sensing for medical diagnostics and security.

The presently available sources of THz radiation can be divided in two categories: optical and electronic. Dye and free-electron lasers belong to the optical group. The former require high-power pumping and are, in principle, monochromatic, with the dye-fixed emission wavelength. The latter cover the whole THz range, but are very large and cumbersome machines. A more practical solution for THz generation is offered by (electrically driven) cascade lasers based on III–V materials. Unfortunately, these devices face severe limitations due to strong phonon absorption. In view of that, strained Si/Ge alloys have recently been considered as an alternative [1]. Yet another approach to THz generation is to use transitions between shallow acceptor levels in Ge and Si [2]. In that case, application of rather large uniaxial stress is frequently required. Most importantly, however, all these solutions require cryogenic temperatures for operation, which is a severe limitation for their applicability.

Optical doping with rare earth (RE) ions has been widely used for development of optical materials and devices. This concerns both insulating hosts and semiconductors, where the availability of an electrical excitation channel appears especially attractive for the development of LED's and electrically driven lasers. RE-doped matrices have also been considered for generation of THz radiation. For that purpose, transitions between individual levels within a single spin-orbit multiplet split by Stark effect of the local crystal-field could be used. For Er-doped crystalline silicon, these studies were conducted at the periphery of investigations for 1.5 μm optoelectronics. Unfortunately, this approach to THz generation turned out to be unsuccessful due

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a large width, and therefore mutual overlap, of emission lines from individual crystal-field split levels of Er^{3+} ions. The large linewidth was traced back to multiplicity of Er-related optical centers simultaneously present in the Er-doped silicon matrix.

Recently, we were able to show that a single type of an Er-related optical center is preferentially formed in Si/Si:Er nanolayers [3,4]. At low temperatures, the photoluminescence spectrum of this center is characterized by a set of ultra-narrow lines with separation of an order of 100 cm^{-1}. With these findings, we have made a major advance towards development of a Si:Er-based THz emitter. In the present study, we have taken advantage of this new finding: using a Si/Si:Er multinanolayer structure, we have explored optical transitions between the well-defined crystal-field split sublevels of the $^4I_{15/2}$ ground state of an Er^{3+} ion.

2. Experimental approach

In the reported study, we have performed pump-probe experiments on a Si:Er structure comprising 400 alternating Si and Si:Er layers. The thickness of the Si:Er layers is 1.7 nm, and the Si spacer layers have a thickness of 2.3 nm. The sample has been grown at a temperature of 520 °C by a sublimation MBE technique [5] on a Cz–Si substrate. The concentration of Er^{3+} and oxygen in the active layers is 2×10^{18} cm^{-3}, and 1.5×10^{19} cm^{-3}, respectively [4]. Past research [3,4] has shown that the Er ions in this sample are preferentially incorporated in a single type of optically active center, designated Er-1, whose emission is characterized by ultra-narrow emission lines.

The measurements have been performed at the Dutch free-electron laser users' facility FELIX. In the experiments, an extended pump-probe scheme was used, which allows for picosecond's resolution in a very broad wavelength range. The investigated sample was placed in an optical gas-flow cryostat. Due to a small energy difference between the levels within the ground state, all the measurements have been performed at cryogenic temperatures.

3. Results and discussion

A high-resolution PL spectrum of a nanolayer structure is presented in Fig. 1. It has been obtained at $T = 4.2$ K and features a series of ultra-narrow lines, thus confirming sharp definition of individual energy levels. The spectrum is due the Er-1 center preferentially formed in a nanolayer structure. In the inset, the small linewidth of a single PL band is illustrated. Splitting within the ground $^4I_{15/2}$ state due to Stark effect of the local crystal-field induces the individual spectral lines.

Fig. 2 illustrates the level scheme of the ground $^4I_{15/2}$ state of the Er-1 center, as derived on basis of combined PL and magneto-PL investigations [4]. The solid black L_j^i arrows denote transitions identified in PL. Possible THz transitions within the ground state are also indicated (dotted arrows). To the best of our knowledge, transitions within the same multiplet of a RE ion have not been observed before, neither in emission nor in absorption. From the scheme, wavelengths of the THz transitions are estimated as 169 μm, 144 μm, 91 μm and 43 μm. In

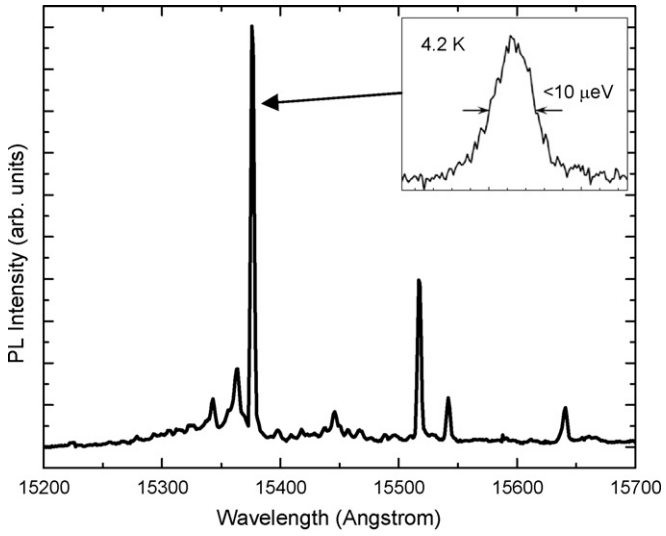

Fig. 1. The Er-related photoluminescence spectrum of the nanolayer structure investigated in the study. The measurement has been taken under continuous-wave excitation with an Ar laser, at $T = 4.2$ K. In the inset: the ultra-small linewidth of the emission lines characteristic for the Er-1 center preferentially formed in Si/Si:Er nanolayers.

this initial search for optical transitions in the Er-related THz domain, we have concentrated on the 43 μm band (the longest dotted arrow).

As the first step of optical exploration, we attempted to measure absorption of our material in the relevant range. This experiment was not conclusive, mostly due to the fact that in the same energy range transitions between levels of shallow donor and acceptor states take place. Since the nanolayer structure used in this study has been grown on a p-type substrate with B acceptor concentration of $[B] \approx 10^{15}$ cm^{-3}, excitations of acceptors obstruct observation of possible Er-related transitions. Based on the concentration argument alone, without any consideration of transition probabilities, one expects a ~7:1 ratio between intensities of Er- and B-related absorption bands.

Nevertheless, one has to remember that in the pump-probe measurements the situation is different: firstly, by the very nature of the measurement, the amplitude of the signal is not linear but quadratic with the incident power. Moreover, in this experiment additional parameter also appears—the lifetime of the excited

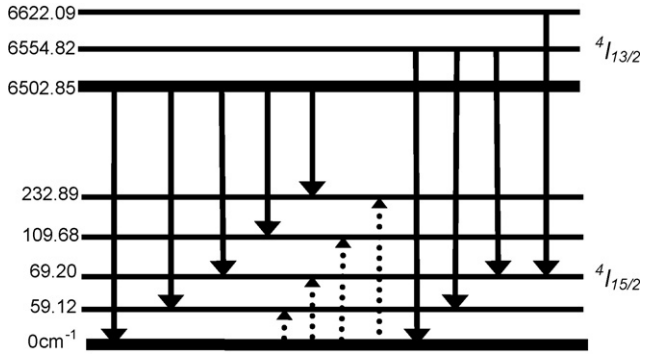

Fig. 2. Energy level scheme of the ground state of the Er-1 center as derived from high-resolution photoluminescence spectroscopy [4].

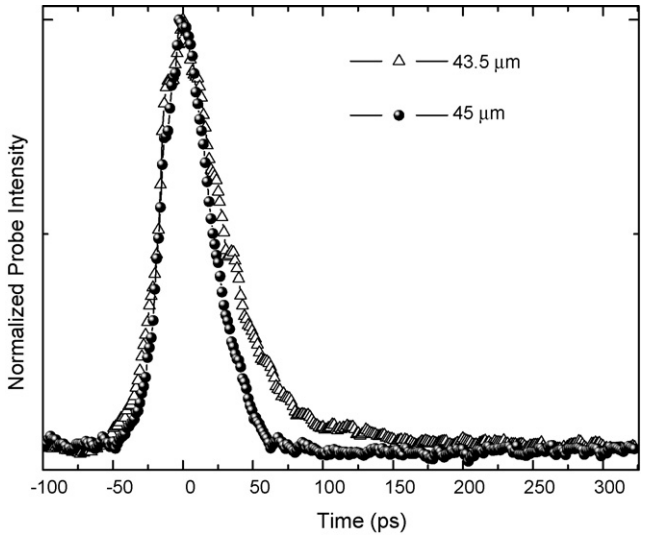

Fig. 3. Comparison of the pump-probe absorption transients as obtained for the free-electron laser set to the "on resonance" and "off resonance" wavelengths of 43.5 μm and 45 μm, respectively.

state participating in the absorption. In case of Er, we are dealing with a forbidden transition, while for effective mass theory (EMT) states of boron the transitions are allowed. However, this becomes less important in view of the fact that in both systems the radiative lifetimes will be much longer than the non-radiative time constant. With the small energy difference, one can expect that the excited state lifetime, governing absorption saturation, will be controlled by non-radiative recombination by phonon emission. For the ∼43 μm transition, the relevant time constant can be estimated as ∼60 ps [6]. Consequently, recombination will be controlled by multiphonon emission. Further, we can expect that the shallow states of boron acceptor are much more effectively coupled to the Si lattice than for an Er dopant—we recall that phonon replicas have never been reported for Er in Si, indicating a relatively small Huang-Rhys factor. In summary, we might expect that in the pump-probe absorption experiment, the Er-related contribution relative to that of boron will be more pronounced than for direct absorption.

This is indeed confirmed by the pilot measurements, which we conducted recently. In Fig. 3, we show an illustration of the observed effect. It compares two absorption transients as measured with the free electron laser tuned on (43.5 μm) and off (45 μm) the expected Er transition; a longer component appearing for the "resonant" wavelength can be distinguished. While these results need to be further confirmed, it is certainly possible that in deed an optically induced transition within the ground-state multiplet of an Er^{3+} ion from an Er-1 center has been observed. This preliminary conclusion is supported by the fact that a similar "slower" transient has also been found for a wavelength of 40.8 μm, which corresponds to one of the $1\,s \rightarrow 2p_{3/2}$ transitions of the B acceptor, identified earlier in direct absorption measurements.

It is necessary to stress that the above findings are only indicative. At this stage, we are, e.g., not able to reliably determine the lifetime of the "longer" component appearing "on-resonance"—neither for erbium nor for boron. We nevertheless note that its value is in the 10–100 ps range, and as such, must be due to the non-radiative recombination by phonon emission, in agreement with our initial estimation [6].

4. Conclusions

Evidence of a radiative transition within the ground state of an Er^{3+} ion in Si has been obtained by pump-probe absorption spectroscopy at 43.5 μm. The characteristic lifetime for the relevant state was determined to be of the order of tens of picoseconds. This is much faster than the estimated radiative decay, indicating that the recombination occurs via phonon emissions.

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