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Observation of Zeeman effect in photoluminescence of Er^{3+} ion imbedded in crystalline silicon

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Abstract

A successful observation of Zeeman effect on Er^{3+}-related photoluminescence in silicon is reported. In a sublimation MBE-grown Si/Si : Er superlattice, a clearly resolved splitting of major spectral components was observed in magnetic fields up to 5.5 T. The Zeeman effect was also investigated for the "hot line" appearing in the spectrum upon temperature increase. Based on the preliminary analysis of the data, the symmetry of the center responsible for the dominant emission line is identified as orthorhombic C_{2v}. Other spectral components originate from at least two more optically active, Er-related centers simultaneously present in the same sample. One of them most probably has cubic T_d symmetry.

Keywords: Photoluminescence; Rare-earth ions; Zeeman effect

1. Introduction

Erbium in various host crystals gives rise to the characteristic emission at $\lambda \approx 1.54\,\mu m$, due to the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of the Er^{3+} ion. Since this particular wavelength is very nearly coincident with the absorption minimum of glass fibers used for telecommunication networks, erbium doping of semiconductors attracts considerable attention. The Si : Er system is of special interest in view of the highly successful and versatile silicon technology. As a result of a continued research effort, Si : Er-based devices, such as light emitting diodes and optical amplifiers, have been successfully developed. In contrast to that, many of the more fundamental aspects of the Si : Er system lack yet satisfactory understanding. In particular, the microstructure of the optically active Er center in silicon is not known.

For Er-doped GaAs, a combination of special growth techniques [1] and spectroscopic investigations [2–4] allowed the microscopic models of two different optically active centers to be proposed. In case of Si : Er, the situation is complicated due to a multiplicity of Er-related centers; more than 100 emission lines have been identified in a high-resolution infrared absorption study on Er-implanted Si [5]. These were assigned to emissions from several, simultaneously present Er-related centers. Moreover, again in contrast to GaAs : Er, but also to, e.g., GaN : Er, individual centers could not be separated by excitation spectroscopy, indicating an (equally) strong lattice coupling of all the species. Prominent formation of cubic center was shown by channeling experiments [6], which identified an isolated Er ion at a tetrahedral interstitial site as the main center generated in crystalline silicon by Er implantation. The channeling studies could not, however, conclude on optical activity of an isolated Er interstitial. Electron paramagnetic resonance has not been, so far, successful in identification of optically active Er-related centers in Si [7]. Also, attempts to observe the Zeeman effect in photolumines-

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cence (PL) of Si : Er were disappointing. Due to the earlier mentioned inhomogeneous character of the line width, application of magnetic field results in broadening and subsequent vanishing of emission lines.

2. Experimental

The recently developed sublimation MBE technique offers new possibilities to develop high quality Si : Er structures. The sample used in the current study has been prepared by this technique and features a quantum structure of 400 interchanged Si and Si : Er layers of a few nanometers thickness. Following the 30 min anneal at 800°C, PL emission from such a sample is at least an order of magnitude more intense than for usual materials prepared by ion implantation. Photoluminescence spectrum at $T = 4.2$ K, depicted in Fig. 1, contains only a few lines of a very small width: $\Delta E \approx 10\,\mu$eV. Upon temperature increase, also shown in the figure, a higher energy "hot line" appears. It corresponds to a transition from a higher lying multiplet of the $^4I_{13/2}$ excited state. Based on the crystal field analysis, predominant formation of a single type of center, labeled Er-1, has been proposed [8,9]. In what follows we will present results of the magneto-optical spectroscopy on this material.

Experiments were performed at liquid-helium temperature using a cw Ar^+-ion laser operating at 514.5 nm for sample excitation. The sample was placed in a split-coil superconducting magnet with optical access (Oxford Instruments Spectromag 4), providing magnetic fields up to 5.5 T. The emerging luminescence was dispersed by a high-resolution 1.5 m F/12 monochromator (Jobin Yvon THR-1500, equipped with a 600 grooves/mm grating blazed at 1.5 μm, and detected with a liquid-nitrogen cooled Ge detector (Edinburgh Instruments)). For polarization measurements, a quarter-lambda plate and a linear polarization filter were used. The Zeeman effect was observed with magnetic field parallel to the $\langle 0\,1\,1 \rangle$ and $\langle 1\,0\,0 \rangle$ crystallographic directions of the sample, the latter one being also the stacking direction of the multilayer structure. The experimental configuration permitted the observation of the luminescence along and perpendicular to the field direction.

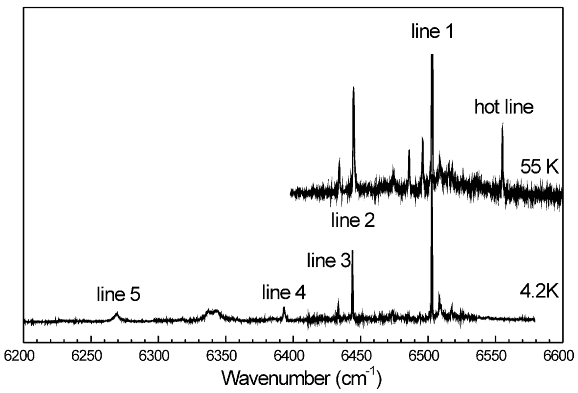

Fig. 1. PL spectrum of the multilayer Si/Si:Er structure used in the current study, as observed at two different temperatures. Individual lines are labeled as used in the text.

3. Results and discussion

In the crystal field of cubic symmetry, the $^4I_{15/2}$ ground and the $^4I_{13/2}$ lowest excited states split in two doublets (Γ_6 and Γ_7) and three quartets (Γ_8), and three doublets (Γ_6 and Γ_7) and two quartets (Γ_8), respectively. The lower symmetry crystal field splits the remaining quartets into doublets. Let us now consider transitions between two Kramers doublets (belonging to a particular configuration of a center of a not defined symmetry). In the magnetic field B, we should observe a splitting of the emission line into four components corresponding to two $\Delta M_J = 0$ (which are linearly polarized), and two $\Delta M_J = \pm 1$ circularly polarized transitions. These will appear at the field dependent energies:

$$h\nu_{1,2}(B) = h\nu(0) \pm \tfrac{1}{2}(G - g)\beta B$$

and

$$h\nu_{3,4}(B) = h\nu(0) \pm \tfrac{1}{2}(G + g)\beta B,$$

where G and g are the effective g-factors of the lower and upper doublet, respectively. In the experiment, we conclude that neither of the lines split off by the magnetic field shows any circular polarization. This indicates that we deal mostly with electric-dipole-type transitions, without spin flips. This is to be expected for Er, since the strong spin–orbit coupling, characteristic for the rare-earth ions, leads to the admixture of different excited configurations (with different L and S quantum numbers but the same J) to the ^4I_J multiplets. In consequence, the linearly polarized $\Delta M_J = 0$ transitions are usually orders of magnitude stronger than the circularly polarized $\Delta M_J = \pm 1$ transitions. In the magnetic field, we should expect to see only transitions without a change of the effective spin.

The transition probabilities for the two lines are equal, since they involve Kramers conjugate states. Any difference in the PL intensities reflects the difference in the population of the magnetic field split states of the upper doublet. In principle, if the spin lattice relaxation time is much shorter than the radiative lifetime, one should be able to determine the g-factor of the upper doublet by comparing the line intensities as a function of B at constant temperature. Unfortunately, at 4.2 K, this is not the case. Studies of the temperature dependence at

4.5 T show that the population of the upper split state decreases with increasing temperature at low temperatures—since the relaxation time is temperature dependent—and only above 30 K the thermalization is really fast enough to obtain any reliable g-factors.

Figs. 2(a) and (b) show the magnetic field-induced splitting of the strongest PL line (marked 1 in the spectrum depicted in Fig. 1) for $B \parallel \langle 1\,0\,0 \rangle$ and $B \parallel \langle 0\,1\,1 \rangle$ configurations. As can be seen, line 1 splits in the magnetic field into three components for $B \parallel \langle 1\,0\,0 \rangle$ and five components for $B \parallel \langle 0\,1\,1 \rangle$. The position of one of them, in both field orientations, almost does not move with the magnetic field strength —for this line, the effective g-factors of the upper and lower state must be almost equal. Assuming that all lines stem from one kind of center and there is no accidental overlap of two PL lines from centers of different symmetry at $B = 0$, we have to consider a low symmetry center with different orientations of the g-tensor axes of the possible configurations with respect to B direction. Naturally, each observed component would then involve transitions between two Kramers doublets.

Cubic T_d or trigonal C_{3v} symmetries cannot explain the observed splitting. It corresponds, however, very well to the orthorhombic $I(C_{2v})$ symmetry type. The number of lines and their relative intensities reflect the expected 2:1 for $\langle 1\,0\,0 \rangle$ and 4:1:1 for $\langle 0\,1\,1 \rangle$. Unfortunately, at this stage of research, due to the lack of experimental data at other field orientations (e.g., $\langle 1\,1\,1 \rangle$) individual g-factors of the upper and lower doublet cannot be uniquely determined.

The C_{2v} symmetry is also consistent with the magnetic field-induced splitting of the "hot line". However, since also in this case we are dealing exclusively with $\Delta M_J = 0$ transitions, individual g-tensors of the involved states cannot be separated. Even assuming that the "hot line" belongs to the same center as line 1, which is a reasonable assumption, we can come up with different sets of g-factors giving an equally good fit. From numerical analysis, we can only conclude that one of the g-tensor values of the lower doublet must be close to zero.

The same C_{2v} symmetry type is also observed for line 4. We conclude, therefore, that lines 1, 4 and the "hot line" could correspond to the same center.

Lines 2 and 3 behave quite differently to lines 1, 4 and the hot line. For B along $\langle 1\,0\,0 \rangle$, line 2 splits into three symmetrical pairs. It cannot stem from transitions between two Kramers doublets of whatever symmetry, and there must be at least one quartet state involved. Since, for $B \parallel \langle 0\,1\,1 \rangle$ similar three pairs of lines are seen, the center seems to have a high symmetry, however, the number of lines is puzzling (for transitions between a cubic quartet and a doublet four lines are expected). At high magnetic fields (above 4 T), the line positions do not depend linearly on B any more and start to curve up to higher energies (especially the lower energy lines). Clearly, there appears to be some interaction between the involved states. One possible explanation is that we deal with a center of cubic symmetry which has a close lying doublet and quartet state in the excited state and a doublet in the ground state (or vice versa). There is no doubt that line 2 does not belong to the same center as line 1.

Line 3 splits into four pairs at $B \parallel \langle 1\,0\,0 \rangle$ and six pairs for $B \parallel \langle 0\,1\,1 \rangle$. It is clearly lower than the cubic site symmetry and must involve transitions between more than two doublets. The situation is clearer than for line

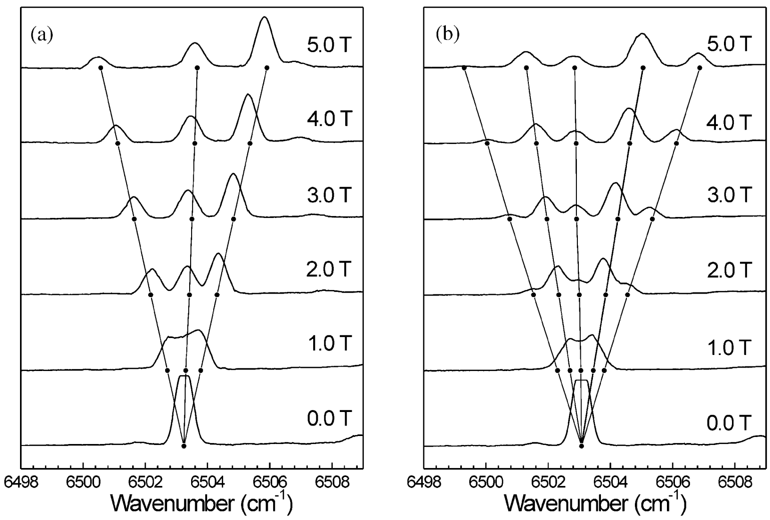

Fig. 2. Magnetic field-induced splitting of line 1 for (a) $B \parallel \langle 1\,0\,0 \rangle$ and (b) $B \parallel \langle 0\,1\,1 \rangle$ configuration.

2. From the analysis of the data, we conclude that there are two doublets in the excited state separated by about $3\,\mathrm{cm}^{-1}$.

4. Conclusions

Based on the preliminary analysis of the Zeeman effect data, we conclude that the individual components of the Er-1 PL spectrum originate from more than one (possibly three) different Er-related optically active centers. The symmetry of the most prominent one is tentatively identified as orthorhombic $\mathrm{I(C_{2v})}$. At least one of the remaining spectral components corresponds to a cubic center ($\mathrm{T_d}$). All the observed PL lines are due to electric dipole-induced transitions without spin flips. Therefore, precise determination of g-tensors at this moment is not possible.

In the continuation of the project, we will investigate Zeeman effect for an arbitrary field orientation, i.e., not along the main directions, where also circularly polarized $\Delta M_J = \pm 1$ transitions should appear. We will look carefully at the intensity changes of individual components at a higher temperature range $T > 30\,\mathrm{K}$, as induced both by temperature and field increase. In this way, we hope to obtain independent information on Zeeman effect for the excited state, allowing for an unambiguous determination of g-tensors. Also, the polarization effects will be carefully investigated.

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