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Study of electronic states of GaAs layers doped with high concentrations of donor and acceptor impurities

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Introduction

Photoluminescence (PL) spectroscopy has been widely used for characterization of semiconductor materials. Although numerous PL studies have been carried out on semiconductor materials up to the present, there is a possibility of drawing significant insights into their impurity and defect states by making the best use of the high sensitivity of this technique. Impurity doped GaAs has been extensively studied by PL spectroscopy[1]. Photoluminescence peaks due to shallow impurity levels which occur at near band-gap energies have been analyzed in details[1]. Another group of PL peaks observed in many earlier studies are those which occur at fairly low energies from heavily impurity doped GaAs[2-11]. Most of these observed peaks have been attributed to complexes of impurity ions and vacancies; the formation of vacancies was explained with their tendency to compensate impurity atoms. In the case of Si doped GaAs, the peaks observed at energies near 1.2eV were attributed to the $\text{Si}_{\text{Ga}}-\text{V}_{\text{Ga}}$ complex[2-9], while those near 1.35eV to the $\text{Si}_{\text{As}}-\text{V}_{\text{As}}$ complex[3, 8, 11]. The existence of these defect complexes in GaAs was suggested by considering similarity to II-VI compounds for which the existence of such defect complexes was confirmed by a number of experiments[2]. In heavily impurity doped semiconductors, however, a variety of other structural changes may occur because of the increasing strength of mutual interactions of impurity ions and the increasing instability of the form of a substitutional solid solution due to incorporation of a high concentration of impurities. There is, therefore, a possibility that PL spectra with signatures of such structure changes may be observed from heavily impurity doped semiconductors.

Objective

The objective of my thesis study is to investigate electronic properties in thick GaAs layers doped with high concentrations of donor and acceptor impurities. The PL spectroscopy has been used as a main method in the study. Specifically the study has been carried out in the following steps. First, the doping processes of high concentrations of donor Si and acceptor Be in GaAs layers by the molecular-beam epitaxy (MBE) growth has been studied along with the characterization of electrical transport properties of the layers. Second, a theoretical method for the analysis of PL spectra of impurity doped GaAs layers has been developed. Finally, experimentally observed PL spectra of GaAs layers doped with high concentrations of Be and Si have been analyzed by using the theoretical method developed in the second step.

Results and Discussion

Doping processes of high concentrations of Be and Si in GaAs layers by MBE at low temperatures with different growth rates were investigated in order to explore the possibility of the low-temperature MBE growth for obtaining novel semiconductor structures. A high concentration of acceptor Be atoms with a hole concentration of $5.65 \times 10^{20} \text{ cm}^{-3}$ in GaAs layer were obtained with a substrate temperature 300°C and a low growth rate $0.03 \mu\text{m/h}$, while an increase in either the substrate temperature or the growth rate resulted in lower hole concentration. This value of the hole concentration is significantly higher than the reported maximum value $2.0 \times 10^{20} \text{ cm}^{-3}$, which was obtained at 450°C with a normal growth rate. In the case of Si doping with low-temperature and low growth rate, Si atoms occupied both donor and acceptor sites, resulting in an electron concentration of $5.59 \times 10^{18} \text{ cm}^{-3}$, which is close to the saturated electron concentration in a Si-doped GaAs layer grown at the normal substrate temperature. In the sample doped with both Be and Si, Si atoms preferentially occupied donor sites with a concentration of $3.35 \times 10^{20} \text{ cm}^{-3}$.

Gallium Arsenide layers doped with high concentrations of Be and Si by MBE are studied by PL spectroscopy. Fig. 1(a) are PL spectra of samples 3a, 3b and 4 which were observed at 4.5K. The doping concentrations of sample 3 and 4 are $5.6 \times 10^{19} \text{ cm}^{-3}$ and $1.1 \times 10^{20} \text{ cm}^{-3}$, respectively. In each spectrum, a sharp peak at 1.27eV corresponds to the 976nm line of the Ar-ion laser which was reflected by the sample surface. A weak peak at 1.49eV is attributed to the recombination of carriers via the carbon acceptor level in the non-doped GaAs buffer layer. A broad peak in each spectrum occurred from the doped layer of a sample. Positions of broad peaks of sample 3a and 3b whose conduction types are different from each other are nearly identical to each other with their maxima at 1.34eV. The energy of the maximum of the peak of sample 4 is 1.32eV. The energy ranges of these broad peaks are similar to those observed from heavily impurity doped GaAs in earlier studies.

As seen in Fig. 1(b), the broad peak shifts to lower energies with an increase in temperature. The magnitude of the shift is comparable to the change in the band-gap of GaAs. The peak energy was found to increase with the increase in temperature in earlier studies. Fig. 1(c) shows the decrease in the peak energy by the annealing, while it was found to increase by the annealing in earlier studies. The doping and growth conditions of the samples and the dependence of the peak energy on the temperature and the annealing shown in Fig. 1(b) and (c) suggest that the origin of the broad peaks observed in the present study is different from that of low energy PL peaks which were observed in earlier studies and attributed to impurity-vacancy complexes. On the basis of these observations, it is suggested that the low energy peaks are attributed to short range ordered arrangements of impurity ions. This possibility is examined by calculations of PL spectra with models of pairs of acceptor and donor delta-doped layers and PL experiments of a superlattice of pairs of Be and Si delta-doped layers.

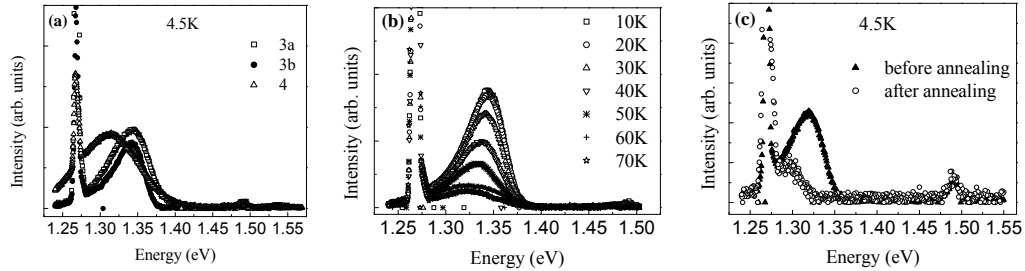


Fig. 1

PL spectra were calculated for a model of the short-range ordered arrangement of Be and Si ions on the basis of the NaCl structure or the CsCl structure both of which are representative crystal structures of ionic crystals; the model is made of a small number of periods of the alternate arrangement of Be and Si delta-doped layers. The calculations were made by using the method developed for theoretical studies on electron states and PL of delta-doped structures[12, 13] for 4.5K with the concentrations of photo-excited electrons and holes $1 \times 10^{15} \text{ cm}^{-3}$.

Fig. 2(a) and (b) are the potential profile and PL spectrum for one pair of Be and Si delta-doped layers with the sheet concentrations in one delta-doped layer is $1 \times 10^{13} \text{ cm}^{-2}$ which corresponds to the sheet concentration of ions in the (111) plane of the perfect NaCl structure made of Be and Si ions with concentrations close to those of sample 3. The PL peak appears at 1.37eV which is lower than the band-gap by the amount of the step-wise energy change. The occurrence of the low energy peak is attributed to the recombination of electrons at one side of the step-wise change of the potential with holes at the other side. The peak position in Fig. 2(b) is close to those of sample 3 in Fig. 1(a). In a short range ordered arrangement, a sheet concentration of ions in a plane corresponding to the (111) plane is expected to be significantly lower than that value. Fig. 2(c) and (d) show results

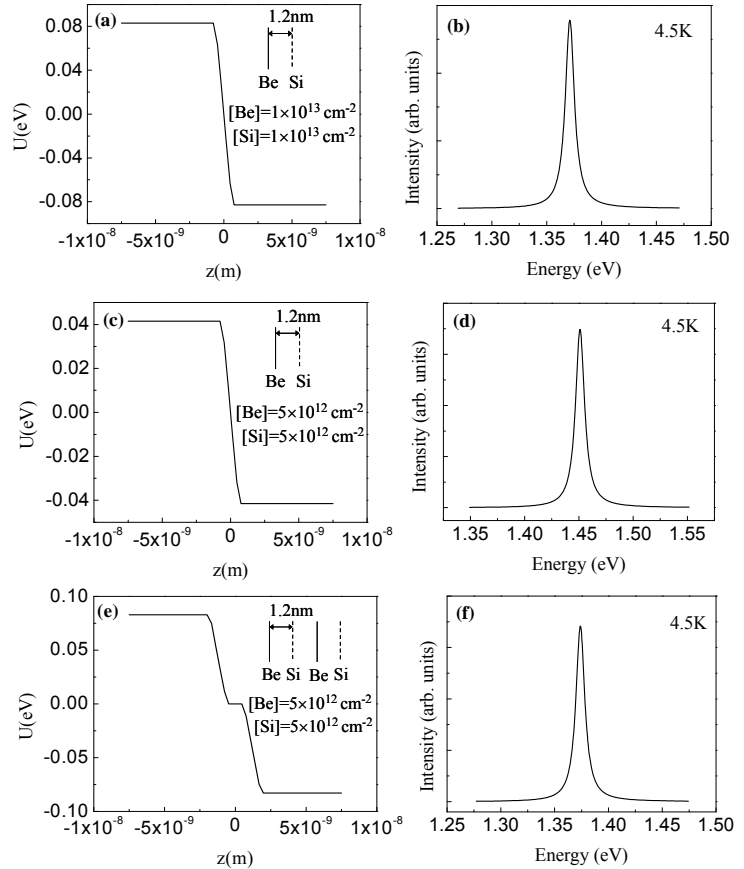


Fig. 2

of the calculations for a pair of Be and Si delta-doped layers with the sheet concentration $5 \times 10^{12} \text{cm}^{-2}$. The peak position moves towards a higher energy than that of Fig. 2(b). Fig. 2(e) and (f) show results of the calculation for two pairs of delta-doped layers with the sheet concentration $5 \times 10^{12} \text{cm}^{-2}$. The change of the potential energy occurs at two steps, resulting in the PL peak energy close to that in Fig. 2(b). These results suggest that short range ordered arrangements of impurity ions in sample 3 and 4 may be represented by a periodic array of several delta-doped layers with sheet concentrations being a fraction of those in the perfect ordered structure.

There has been, however, no earlier report of such an observation of a PL peak from impurity doped semiconductors. In addition, there is no other experimental observation which indicates the formation of a short-range ordered arrangement of Be and Si ions in the present samples. We, therefore, grew a superlattice of Be and Si pair delta-doped layers and observed its PL spectrum in order to examine the above-mentioned possibility. Fig. 3(a) shows schematically the structure of the superlattice. The growth of a pair of Be and Si delta-doped layers at 400°C was repeated 180 times in order to obtain sufficient PL intensities from delta-doped layers. The sheet impurity concentration of each delta-doped layer was intended to be $2.4 \times 10^{13} \text{cm}^{-2}$, but the concentration of Si delta-doped layers may have been different from this value. Fig. 3(b) is an observed PL spectrum which shows a broad peak at 1.32eV. Fig. 3(c) is the calculated PL spectrum which shows the peak at an energy close to the observed one. For the calculation, a pair of Be and Si delta-doped layers with sheet concentrations of $2.4 \times 10^{13} \text{cm}^{-2}$ and $2.1 \times 10^{13} \text{cm}^{-2}$, respectively was used as a model. Although a superlattice of a pair of delta-doped layers is different from a short range ordered arrangement of impurity ions, the similarity of observed PL peaks in Fig. 1(a) and Fig. 3(b) suggests that the latter PL peaks results from the short range ordered arrangement.

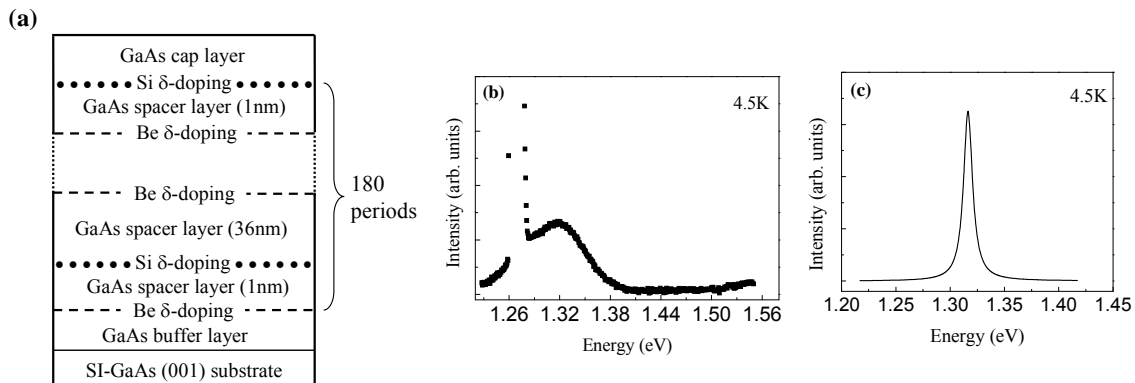


Fig. 3

Significance of the study

The doping processes of high concentrations of donor Si and acceptor Be in GaAs layers by MBE at low temperatures with different growth rates were investigated along with the electrical transport properties of the layers in order to clarify the microscopic processes which determined their maximum concentrations in the low-temperature growth. PL peaks from doped layers were observed at energies significantly lower than the band-gap of GaAs. The analysis of PL spectrum with theoretical calculations has shown that the Be and Si ions have formed short range ordered arrangements on the nano-meter scale caused by their Coulomb interactions. The present study, therefore, has demonstrated the novel capability of PL spectroscopy for the analysis of impurity distributions in semiconductors in the nano-meter scale.

Another significance of the present study is the new finding of electronic structures of donor and acceptor doped semiconductors. Semiconductors with high concentrations of donor and acceptor impurities have been studied from the fundamental point of view with respect to the Anderson localization of carriers due to a random ionic potential[14]. The present study has revealed a quite different picture of carrier distributions in these semiconductors. Such distributions of electrons and holes results in an optical property which is highly different from that in ordinary intrinsic semiconductors. The interpretation of electron transport properties in these donor and acceptor doped semiconductors should also be changed from that based on the random ionic potential. These new findings are, therefore, expected to lead to researches which are aimed exploration of new possibilities of donor and acceptor doped semiconductors from both fundamental and application-oriented points of view.

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