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Identification of an isolated arsenic antisite defect in GaAsBi

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Optically detected magnetic resonance and photoluminescence spectroscopy are employed to study grown-in defects in GaAs_{0.985}Bi_{0.015} epilayers grown by molecular beam epitaxy. The dominant paramagnetic defect is identified as an isolated arsenic antisite, As_{Ga}, with an electron g-factor of 2.03 ± 0.01 and an isotropic hyperfine interaction constant $A = (900 \pm 20) \times 10^{-4} \text{ cm}^{-1}$. The defect is found to be preferably incorporated during the growth at the lowest growth temperature of 270 °C, but its formation can be suppressed upon increasing growth temperature to 315 °C. The As_{Ga} concentration is also reduced after post-growth rapid thermal annealing at 600 °C.

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Incorporation of bismuth (Bi) in dilute quantities in GaAs has been shown to lead to several intriguing, potentially desirable changes in the fundamental properties of GaAs. The most articulated modifications stem from the interaction of Bi 6p bonding orbitals with the valence band (VB) maximum of GaAs, resulting in a large bowing effect in both the bandgap energy^{1–7} and the spin-orbit (SO) splitting.^{4,6,7} For example, addition of 1% of Bi results in a decrease of the bandgap energy by 0.09 eV,^{5,7} while the SO splitting increases by 0.06 eV.⁴ These changes indicate that GaAsBi belongs to a class of highly perturbed alloys.

The giant bandgap bowing in GaAsBi opens possibilities for the design of highly efficient optoelectronic devices,⁷ transistors with a low power consumption, multi-junction solar cells, etc. For example, it is expected that temperature-insensitive 1.3 and 1.55 μm laser diodes, which are important light sources for fiber-optic communications, can be realized by using GaNAsBi alloys with a low percentage of N.⁸ GaAsBi is also an attractive material for the base region in heterojunction bipolar transistors (HBTs), since a small reduction in the bandgap of the base material can significantly reduce the power consumption, which is critical for portable wireless devices. In addition, as bismuth is the heaviest non-radioactive element, alloying with it creates an unusually large SO-splitting⁴ due to a large relativistic correction to the host electronic band structure. The possibility of tailoring the SO splitting is highly desirable for spintronic devices where strong coupling between the spin and the orbital angular momentum of electrons could enable spin manipulation by an electric field. Furthermore, the combined effect of the decreased bandgap energy and the increased SO-splitting results in a larger SO splitting than the bandgap energy at [Bi] = 10.5%.⁶ This effect has been suggested to lead to quenching of undesirable Auger recombination in GaAsBi as the energy released during recombination between a conduction electron and a heavy hole (hh) becomes insufficient for the Auger excitation of a hole between the hh- and the SO split-off valence subbands. This

would be highly beneficial for the design of high-efficiency lasers.

The aforementioned intriguing and useful properties of GaAsBi are caused by the large size and low electronegativity of Bi as compared with As. The difference in sizes, however, also imposes a requirement for non-equilibrium growth conditions, e.g., low growth temperatures below 400 °C, in order to achieve Bi incorporation.^{1,7,9} Such non-equilibrium growth is known to favor formation of a large amount of native defects, such as arsenic antisites (As_{Ga})¹⁰ in GaAs grown at low temperatures (LT) by molecular beam epitaxy (MBE), or gallium interstitials (Ga_i) in GaNAs^{11–17} and GaNP^{18–21} alloys. From this point of view, abundant defects are expected to be introduced during the growth of GaAsBi. On the other hand, Bi is known to be a good surfactant, improving²² the surface morphology and photoluminescence (PL) properties of, e.g., GaNAs layers grown at 460 °C. These results have been interpreted as an indication that the surfactant tends to reduce the density of electronically active defects. Thus, surface kinetics during growth might be altered by Bi.

At present, research devoted to dilute bismides is still at an early stage of material development when optimization of growth parameters and control of grown-in defects are crucial. By analogy to other highly mismatched semiconductors (as well as LT-MBE GaAs), it is expected that effects of defects on optical and electrical properties should be very strongly manifested. Indeed, recent DLTS measurements^{23–25} have revealed the presence of a number of carrier traps in GaAsBi alloys. Bi_{Ga} antisite defects were also reported²⁶ in GaAs lightly doped with Bi. An improved understanding of the microscopic structure of major grown-in defects and impurities along with their influence on physical properties of the GaAsBi alloy is critically needed in order to control them and to fully explore the potential of dilute bismides for device applications.

The aims of the present work are (a) to closely examine and unambiguously identify important grown-in defects in GaAs_{1–x}Bi_x, (b) to obtain information about the role of defects in carrier recombination processes, and finally, (c) to evaluate the effects of growth temperature and post-growth thermal annealing on the defect formation. PL, PL excitation

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FIG. 1. XRD spectra from the as-grown $\text{GaAs}_{0.985}\text{Bi}_{0.015}$ samples grown at different temperatures (T_g).

(PLE) and optically detected magnetic resonance techniques (ODMR) will be employed for these purposes.

Two $\text{GaAs}_{0.985}\text{Bi}_{0.015}$ epilayers used in this study were grown at two different growth temperatures (270 °C and 315 °C) on a semi-insulating GaAs (001) substrate by solid-source MBE. The growth started with a 120 nm-thick GaAs buffer layer grown at normal conditions (580 °C and a high As overpressure), and was followed by a GaAsBi epilayer (230–250 nm in thickness) grown at a lower temperature with an As/Ga flux ratio close to one as required for efficient Bi incorporation.² Additionally, a reference GaAs epilayer was grown under similar conditions as described above but without Bi incorporation and with an As/Ga flux ratio about twice as high as for the GaAsBi samples. The Bi concentration was 1.5% in both GaAsBi samples, as determined by high resolution x-ray diffraction measurements, using the literature value of 6.33 Å for the GaBi lattice constant.² We should note, however, that in addition to Bi incorporation, As_{Ga} antisite defects can contribute to the expansion of the lattice constant thus causing overestimation in the Bi composition by X-Ray diffraction (XRD).²⁷ Representative XRD spectra for the as-grown GaAsBi samples are shown in Fig. 1, measured with a triple-axis X-ray diffractometer from (004) planes using the Cu-K α radiation. The appearance of the well-defined thickness fringes is indicative of good crystal quality and well-defined interfaces. Detailed study of structural properties of similar (but not identical) GaAsBi layers is reported in Ref. 28. One piece from each GaAsBi sample was then treated by rapid thermal annealing (RTA) at 600 °C for 60 s in a N_2 ambient. PL and ODMR measurements were performed at 5 K using either the 915-nm line of a solid state laser or the 751-nm line of a Ti:Sapphire laser as an excitation source. The PL signals were dispersed by a 1-m double grating monochromator and were detected by a liquid nitrogen-cooled Ge detector. ODMR signals were measured at both X-band (9.213 GHz) and Q-band (33.87 GHz) as spin-resonance-induced changes of the PL intensity (detected by a Ge detector) utilizing the lock-in technique in phase with an amplitude-modulated microwave field at a frequency of ~ 3 kHz.

Figure 2(a) shows the effects of growth temperature and annealing on the PL spectra of the $\text{GaAs}_{0.985}\text{Bi}_{0.015}$ alloy measured at 5 K. In the spectral region of 1.0–1.28 eV,

FIG. 2. (a) Representative PL spectra measured at 5 K from the studied LT $\text{GaAs}_{0.985}\text{Bi}_{0.015}$ epilayers. The spectra are taken with an excitation photon energy exceeding the GaAs bandgap. (b) Isotropic ODMR spectra obtained at Q-band (33.87 GHz) by monitoring the PL emissions shown in (a). The ODMR signals are negative and are shown as positive for easy viewing. The ODMR spectra were measured at 5 K with the excitation photon energy of 1.35 eV.

GaAsBi exhibits a strong, broad PL band centered at ~ 1.15 eV, which is not present in the Bi-free reference structure. This PL emission is tentatively ascribed to recombination of localized excitons. This conclusion is supported by following experimental observations: (i) a blueshift of the PL peak position with increasing optical excitation power, see Fig. 3(a); (ii) an increase of a temperature-induced redshift of the PL peak position with lower excitation powers (Fig. 3(b)); and (iii) a large Stokes shift of 0.3 eV, indicative of strong localization. The origin of the localized excitons could be excitons either localized at defect states or bound to the band tail states extending from the band edge due to a strong fluctuation of the bandgap as a result of local variations in the alloy composition. The alloy band gap value deduced from the PLE cut-off is 1.28 eV.

Typical ODMR spectra obtained by monitoring the 1.15 eV emission are presented in Fig. 2(b). For the epilayer grown at 270 °C, a strong ODMR signal is observed and contains four equidistant lines. The intensity of this signal is dramatically reduced in the sample grown at 315 °C, which indicates a significantly lower defect concentration in the alloy grown at the higher temperature. As the same substrate material was used and the GaAs buffer layers were grown under identical conditions in both samples, we can rule out the possibility that the observed ODMR signal originates from the substrate or the buffer layers. Otherwise the same ODMR signal would have been observed in both samples regardless of the growth temperatures of the GaAsBi epilayers. We can also exclude the possibility that the ODMR signal originates from a defect at the GaAs/GaAsBi interface because a defect at an interface should exhibit a reduced symmetry giving rise to an anisotropic spin-resonance signal, such as the P_b defects at Si/SiO₂ interface²⁹ and the DD1

FIG. 3. (a) PL peak position as a function of excitation power. The filled circles are the experimentally determined values at 5 K. The line is a guide to the eye. (b) PL peak position as a function of measurement temperature at two excitation power levels. The filled circles and squares are the experimentally determined values at 5 K. (c) PL and PLE spectra obtained at 5 K. The vertical arrow indicates the detection energy monitored in PLE. The tilted arrow indicates the cut-off energy of the PLE. All data are from the as-grown $\text{GaAs}_{0.985}\text{Bi}_{0.015}$ epilayer grown at 270°C .

defect at GaP/GaN interface,³⁰ which is in contradiction to the isotropic ODMR signal observed here. Thus, we can conclude that the ODMR signal must originate from a defect situated in the GaAsBi epilayer. The observed ODMR signal is negative, i.e., corresponding to a decrease in the monitored PL intensity upon the spin-resonant transitions. It is shown as positive in Fig. 2(b) merely for easy viewing. The observed decrease in the intensity of the monitored PL under the spin-resonance condition implies that the PL is most likely not directly related to the defect involved in the spin resonance. Instead, it is a result of competing carrier recombination via a defect of a different origin from the one giving rise to the PL.^{31,32} Carrier recombination via the former accelerated by the spin-resonance transitions can thus lead to a corresponding decrease in the radiative carrier recombination of the latter (i.e., the 1.15 eV PL band).

To analyze the quadruplet ODMR spectrum, the following spin Hamiltonian is used in order to calculate the energy levels of the spin system at the defect and the expected magnetic-resonance field positions: $H = \mu_B g \mathbf{B} \cdot \mathbf{S} + A \mathbf{S} \cdot \mathbf{I}$. Here, μ_B is the Bohr magneton, $\mathbf{B}$ is the external magnetic field, and A is the central hyperfine (hf) parameter that describes the coupling of the electron spin $\mathbf{S}$ with the nuclear spin $\mathbf{I}$ of the defect. The electronic g -factor and the A parameter are scalars here, since all observed ODMR signals are isotropic. As shown in Figs. 4(a) and 4(b), the quadruplet structure can be reproduced assuming a paramagnetic defect center with a localized electron spin $S = 1/2$ that is strongly coupled by the hf interaction to a nuclear spin of $I = 3/2$ with 100% natural abundance. The only chemical element that satisfies these requirements is arsenic. Therefore, the obtained ODMR results provide an unambiguous experimental proof that the monitored paramagnetic defect involves an As atom.

FIG. 4. Representative ODMR spectra measured at 5 K in (a) X-band and (b) Q-band from the $\text{GaAs}_{0.985}\text{Bi}_{0.015}$ epilayer grown at 270°C . The upper-most curves in (a) and (b) are the experimental spectra measured by monitoring the total intensity of the PL emissions in the 950–1200 nm spectral range. The simulated ODMR spectra from the As_{Ga} defects are shown by the lowest curves. They were obtained by using following spin-Hamiltonian parameters: $g = 2.03 \pm 0.01$ and $A = (900 \pm 20) \times 10^{-4} \text{ cm}^{-1}$ and by assuming a Gaussian lineshape with a linewidth of 35 mT. The ODMR signals are negative but they are shown as positive for easy viewing.

A best fit to the experimental data are obtained by using the following spin-Hamiltonian parameters: $g = 2.03 \pm 0.01$ and $A = (900 \pm 20) \times 10^{-4} \text{ cm}^{-1}$. The simulated ODMR curves by using these parameters are shown in Figs. 4(a) and 4(b) for the X- and Q-band, respectively. The good agreement between the simulations and the experimental results justifies the reliability of the obtained fitting parameters.

A comparison of the determined spin Hamiltonian parameters with the previously reported magnetic-resonance signatures of As-containing defects in GaAs shows that the defect in fact belongs to the most thoroughly investigated family of intrinsic defects: As antisites (As_{Ga}). The formation of these defects in the studied GaAsBi is probably not surprising, as low growth temperatures and As overpressure during the MBE growth are known³³ to result in the incorporation of excess As in the GaAs lattice leading to an abundance of As antisites in LT GaAs. Although the As pressure is reduced during the GaAsBi growth in order to ensure Bi incorporation, both Bi and As atoms compete for the same lattice sites which may further promote the As_{Ga} formation. Our results also show that the defect formation is largely suppressed with increasing growth temperature from 270°C to 315°C . In addition, the As_{Ga} -related ODMR signal is completely quenched after an RTA treatment at 600°C for 60 s. This is in agreement with the previous studies^{34–36} of As_{Ga} in LT-GaAs, where thermal annealing at similar temperatures has been found to reduce defect concentrations, due to clustering of As atoms.

It is difficult to exactly identify the nearest neighbors of the As_{Ga} atom since ligand hyperfine interactions are not resolved in the measured ODMR spectra. We note, however, that the defects that belong to the As_{Ga} family in GaAs differ in the hf interaction strength A , which ranges^{34–43} from 680×10^{-4} to $900 \times 10^{-4} \text{ cm}^{-1}$. The largest value is expected for an isolated As_{Ga} defect, whereas a reduction in A has been taken as evidence for complex formation that leads to a re-distribution of the electron wavefunction from As_{Ga} to its neighboring atoms. Thus, judging from the strength of the hf interaction, we conclude that the defect observed in the GaAsBi alloy grown at 270°C is most likely an isolated

As_{Ga}^+ center with four As neighbors. This defect configuration is also typical for LT GaAs.^{34–39} The fact that the spin Hamiltonian parameters remain unaffected by the presence of 1.5% of Bi in the GaAs lattice is a result of the strong localization of the electron wavefunction at the As antisite which is known to be a deep donor. Furthermore, the linewidth (35 mT) of the magnetic resonance lines observed here is identical to that reported previously for As antisites in GaAs.^{34–43} As the linewidth is determined by the hyperfine interaction with the ligand atoms surrounding the As_{Ga} , this finding is thereby indicative of identical local surrounding of As_{Ga} in both GaAs and GaAsBi, i.e., Bi is not one of the nearest neighbors of the As_{Ga} in GaAsBi studied here. Previous ESR studies²⁶ of Czochralski-grown GaAs lightly doped with Bi revealed that a substantial fraction (about 10%) of Bi dopants occupied the Ga site, forming Bi_{Ga} antisite defects. Despite considerable efforts, however, we found no experimental evidence for such behavior in the studied GaAsBi alloys.

In conclusion, we have conducted a detailed study of grown-in defects in MBE-grown GaAsBi epilayers. A solid experimental proof is provided that the dominant paramagnetic defect, which is formed in the GaAsBi epilayer grown at 270 °C, contains an As antisite atom at its core. Based on the comparison of the deduced spin-Hamiltonian parameters for As_{Ga} in GaAsBi with those known for As_{Ga} -related defects in GaAs, the defect configuration is concluded to be an isolated As_{Ga}^+ center surrounded by four As neighbors. The defect acts as a competing recombination center (judging from the negative ODMR signal) and could, therefore, be harmful to the performance of optoelectronic devices based on the GaAsBi alloy. It is shown to be preferably incorporated during the growth at the lowest growth temperature, and its formation is suppressed upon increasing growth temperature from 270 to 315 °C. Post growth rapid thermal annealing at 600 °C was found to completely quench the As_{Ga} ODMR signal. The dependence of the defect formation on growth temperature and post-growth thermal annealing is in line with the previously reported properties of As_{Ga} in LT-GaAs.

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