We report on the structural and electrical characterizations of MnAs/GaAs hybrid nanowires fabricated by combining selective-area metal–organic vapor phase epitaxy of undoped GaAs nanowires and endotaxial nanoclustering of MnAs. As a result of endotaxy, MnAs nanoclusters are typically embedded in the six ridges of hexagonal GaAs nanowires. However, the MnAs nanoclusters are formed not only at the six ridges, but on the surfaces of six (011) facets of hexagonal GaAs nanowires, when we decrease the growth temperature from 600 to 400°C. From the cross-sectional characterizations by transmission electron microscopy, the size and density of the MnAs nanoclusters formed at the top part of the GaAs nanowires are much larger than those formed at the bottom part of the nanowires. Current and voltage characteristics of MnAs/GaAs hybrid nanowires are investigated using two-terminal device structures of free-standing hybrid nanowires. The hybrid nanowires formed on Zn-doped p-type GaAs (111)B substrates show ohmic characteristics, while those formed on Si-doped n-type GaAs (111)B substrates show clear rectifying characteristics. The hybrid nanowires show p-type conductivity possibly owing to the formation of p-type GaAs layers near the nanowire surfaces caused by the thermal diffusion of Mn atoms into GaAs nanowires during the endotaxial nanoclustering of MnAs.

On the other hand, the heteroepitaxy of ferromagnetic and III–V compound semiconducting layers (FM III–V hybrids) has intensively been investigated in the research field of nanospintronic devices, such as magnetic sensors and spin-polarized light-emitting diodes fabricated using NWs. We believe, in particular, that NWs are a promising candidate among the FM III–V hybrids for future magnetonanoelectronic devices because it has been reported that they show huge magnetoresistance (MR) effects. However, technological applications of the granular hybrid structures fabricated by a conventional technique, that is, molecular beam epitaxy under extremely low growth temperature conditions and a subsequent top-down-type etching method, are mainly restricted to macroscopic devices because of the random distribution of NCs in the host materials, e.g., the randomness in the mean distance between NCs and in NC size and shape, which strongly affects the MR effects or magnetotransport properties and possibly leads to statistical fluctuations of device characteristics. This might be a serious problem that we have to solve in future miniaturized devices. Recently, NWs fabricated using diluted magnetic materials or FM III–V hybrids have also been realized by the conventional VLS method and thermal annealing after the deposition or the ion beam implantation of Mn nanoparticles onto the VLS-grown NWs for nanospintronic device applications using NWs.

Our SA-MOVPE, which is a bottom-up-type fabrication method in combination with lithographical techniques, on the other hand, has successfully demonstrated position and size controllability in the formation of ferromagnetic MnAs NCs, in addition to semiconductor NWs, on GaAs (111)B substrates. We found in our previous studies of the ferromagnetic NCs on semiconductor wafers that ordered arrays of the chain structures comprising elongated MnAs NCs strongly affected the MR effects of the applied currents and showed angle-dependent MR. These results showed the possibility of applying the hybrid NWs to magnetonanoelectronic devices, such as magnetic sensors and spin-polarized light-emitting diodes fabricated using NWs. We believe that NWs are promising candidates for magnetic sensor device applications, to enhance the angle-dependent MR effects of the applied current in the NW channels, because of their aspect ratio being possibly larger than 100 and the ordered arrays of ferromagnetic NCs around the NW channels. We have successfully demonstrated the fabrication of hybrid structures between MnAs...
NCs and GaAs NWs selectively grown on GaAs (111)B substrates as a first prototype structure. In this paper, therefore, in addition to the detailed cross-sectional observation results, we present the results of the electrical characterization of the hybrid structures, since there is no information on the conductivity of the MnAs/GaAs hybrid NWs fabricated by combining SA-MOVPE of GaAs NWs and endotaxial nanoclustering of MnAs. Fundamental MOVPE growth condition dependence of the formation of MnAs NCs on the GaAs NW array templates will also be discussed.

2. Experimental Procedure

The templates of the typical GaAs NW arrays on GaAs (111)B substrates were fabricated by our typical SA-MOVPE process, which has been described in detail elsewhere. We first fabricated the initial circular open-MOVPE process, which has been described in detail (111)B substrates were fabricated by our typical SA-MOVPE process, which has been described in detail (111)B substrates were fabricated by our typical SA-MOVPE process, which has been described in detail elsewhere.

We supplied the gases of (CH\(_3\))\(_2\)Ga and 20% AsH\(_3\) diluted in H\(_2\), as well as, respectively, in the current experiments. Conventional organo-metallic and hydride sources, such as (CH\(_3\))\(_3\)Ga and 20% AsH\(_3\) diluted in H\(_2\), were used as the group III and group V sources for all the growth experiments, respectively.

For the MnAs NC growth after the growth of undoped GaAs NW templates, we utilized the phenomenon of “endotaxy” of MnAs in GaAs. We have observed that single-crystal MnAs NCs were grown in GaInAs (111)A layers even without any supply of an AsH\(_3\) source as a result of endotaxy. This is the key technique for forming MnAs NCs “into” undoped GaAs NWs, as demonstrated in our previous study. The detailed explanation for the phenomenon of endotaxy was reported elsewhere. We supplied the gases of (CH\(_3\))\(_3\)Ga and 20% AsH\(_3\) diluted in H\(_2\), as well as, respectively, in the current experiments. Conventional organo-metallic and hydride sources, such as (CH\(_3\))\(_3\)Ga and 20% AsH\(_3\) diluted in H\(_2\), were used as the group III and group V sources for all the growth experiments, respectively.

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The growth temperature \(T_g\) and the V/III ratio for the GaAs NW growth were 750 °C and about 180, respectively. Conventional organo-metallic and hydride sources, such as (CH\(_3\))\(_3\)Ga and 20% AsH\(_3\) diluted in H\(_2\), were used as the group III and group V sources for all the growth experiments, respectively.

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and density of the NCs were strongly influenced by the gas supply conditions in this purging process. Therefore, we introduced, in the current work, only the hydrogen, H$_2$, carrier gas in the MOVPE reactor during the decrease in $T_g$ after the endotaxy of the MnAs NCs. In Fig. 2, the distance between the NWs, $a$, was set to 3.0 $\mu$m for all the observed samples. The size of the MnAs NCs was markedly increased, as shown in Figs. 2(a) and 2(b), when we decreased $T_g$ for the endotaxy of the MnAs NCs from 600 to 500 $^\circ$C. With decreasing $T_g$ for the MnAs NC formation to 400 $^\circ$C, on the other hand, the size of the MnAs NCs markedly decreased, and, in addition, the NCs were formed not only on the six ridges, which are the areas between two of the $\{0\overline{2}11\}$ facets, of hexagonal GaAs NWs from the top to the bottom part of the NWs, but also on the $\{0\overline{1}1\}$ facet surfaces, as shown in Fig. 2(c).

Next, we observed the cross-sectional images of the hybrid structures between MnAs NCs and GaAs NWs from the top to the bottom part of the NWs by TEM. The growth conditions for the NCs and the atmospheric conditions after the NC growth for the observed structure were the same as those for the sample shown in Fig. 2(a), but the distance between the NWs, $a$, was 0.5 $\mu$m. Figures 3(a) and 3(b) show a typically observed image of the whole structure of a hybrid NW and the schematic illustrations and the crystal orientations for the cross-sectional observations, respectively. The height and diameter of the observed structure in Fig. 3(a) were estimated to be 1.7 $\mu$m and 210 nm,
respectively. We confirmed from Fig. 3(a) that the hybrid structures had the same diameter, 210 nm, over the whole structure from the top to the bottom, by which we can nearly ensure that the \{011\} facets were formed as side walls of hexagonal GaAs NWs from the top to the bottom. From the cross-sectional TEM images at the top and bottom parts of the NWs in Figs. 3(c) and 3(d), respectively, we estimated the average width and depth of the NCs in the NWs to be about 14.8 and 6.0 nm at the top part of the NWs, and about 10.7 and 4.0 nm at the bottom one, respectively. The average density of the NCs, which was defined as the number of NCs per ridge of the NWs, i.e., /\mu m, was also estimated to be about 19.6 at the top part of the NWs and about 15.5 at the bottom one. To estimate solid compositions in the NCs and NWs, in addition, analysis by EDX spectroscopy was conducted for the three elements Mn, As, and Ga in the NCs and NWs. Here, for the EDX analyses, the atomic percentages of the three elements were estimated after eliminating the possible external contaminations by carbon, oxygen, and silicon atoms presumably during the sample preparation processes and originating from the material of the sample holders for the TEM observations. The solid compositions (at. %) of Mn, As, and Ga in one of the NCs were estimated to be approximately 44, 50, and 6.0%, and, in another NC on the same NW, to be approximately 33, 56, and 11%, respectively. In the middle of the NW, we detected no Mn element, but approximately 48% of As and 52% of Ga elements. Therefore, a small amount of Ga atoms from 6.0 to 11% was possibly included in the MnAs NCs presumably owing to the redistribution of the host materials as a result of the endotaxy of MnAs NCs in the host GaAs NWs, but we confirmed in our previous study\(^{37}\) that all the crystal structures of the MnAs or Mn(Ga)As NCs and the GaAs NWs observed were of the hexagonal NiAs- and cubic zinc-blende types, as determined from electron-beam diffraction measurements, respectively. Taking together all the findings obtained in these growth experiments and structural characterizations, we discuss the possible explanation for the growth behaviors of MnAs NCs on GaAs NWs. In the SA-MOVPE growth process, there possibly are three major contributions in the diffusion of source molecules (or species): the first is the diffusion from the vapor phase, the second is the surface diffusion of adatoms on SiO\(_2\)-mask surfaces, and the final is the diffusion between the surfaces of crystal facets. In the case of the MnAs NCs grown at 400 °C, as shown in Fig. 2, we observed NC formation not only on the six ridges of hexagonal GaAs NWs, but also on the six \{011\} facet surfaces. This experimental result presumably shows the decrease in the diffusion rate of Mn adatoms between the \{011\} facets. In addition, the diffusion of Mn atoms from the vapor phase was much more predominant in our experiments than the surface diffusion of Mn adatoms on the SiO\(_2\)-mask surfaces, because the size and density of the NCs at the top part of the NWs were larger than those at the bottom part of the NWs, as observed in Fig. 3. It appears that these results are consistent with the growth behaviors of the NCs in terms of the different growth parameters discussed in our previous study.\(^{37}\)

### 3.2 Electrical characterization of hybrid structures between MnAs NCs and GaAs NWs

Next, to evaluate electrical properties of hybrid structures between MnAs NCs and GaAs NWs, we first developed a device process. Figure 4 shows schematic illustrations for the developed fabrication procedure for the two-terminal device structures. Corresponding images taken by SEM for each of the steps of the device process are also shown in Fig. 4. We have modified the developed procedure for the fabrication of nanowire light-emitting diodes reported in our previous study.\(^{40}\) After the growth of hybrid structures between MnAs NCs and undoped GaAs NWs [Fig. 4(a)], the

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**Fig. 4** (Color online) Process for the electrical characterization and fabrication of two-terminal devices with corresponding images taken by SEM and schematic illustrations at each of the steps.
space between the hybrid structures was filled with polymer resin, that is, benzocyclobutene (BCB) in this experiment, by spin coating, to put an electrode on the top part of hybrid structures and ensure the insulation between two electrodes for the electrical measurements. After the spin coating of BCB, the samples were annealed at 300 °C for 15 min for the solidification of BCB [Fig. 4(b)]. Subsequently, the top portion of the hybrid structures overlaid with an excessive amount of BCB was exposed by reactive ion etching (RIE) using the gases of CF₄ and O₂. The height of the exposed hybrid structures after the RIE was estimated to be about 100 nm [Fig. 4(c)]. Next, after the photolithography process to form top electrode patterns for each of the hybrid structure arrays on the substrates, 30-nm-thick Cr and 50-nm-thick Au film electrodes were deposited onto the exposed hybrid structures by evaporation in vacuum, and then, lift-off was carried out to remove the electrodes outside the areas with hybrid structure arrays. Finally, the back-side electrodes were formed on the substrates. For Zn-doped p-type GaAs (111)B substrates, 30-nm-thick Cr and 50-nm-thick Au film electrodes were deposited, and, for Si-doped n-type ones, composite film electrodes of Au/Ge/Ni/Au, whose thicknesses were 25, 20, 15, and 30 nm, respectively, were deposited by evaporation in vacuum [Fig. 4(d)].

Using the developed device process in Fig. 4, we fabricated two-terminal device structures for evaluating electrical properties of hybrid structures between MnAs NCs and undoped GaAs NWs. The growth temperature T₉ for the endotaxy of MnAs NCs was 600 °C for all the samples used in the electrical characterizations. Figure 5(a) shows a summary of the typical I–V characteristics of the hybrid structures, in which the distances between the NWs, a, were 0.5, 1.0, and 3.0 μm, fabricated on Zn-doped p-type GaAs (111)B substrates. For the sample with a = 1.0 μm, the diameter of the GaAs NWs was estimated to be about 150 nm, and approximately 8,500 NWs were included under one electrode of the device. On the p-type substrates, I–V curves obtained in the current work showed approximately linear characteristics, i.e., ohmic behavior, for all the samples. The resistance of the samples with a = 0.5 and 1.0 μm was estimated to be about 1.0 and 6.4 MΩ at room temperature, respectively. In the case of the sample with a = 3.0 μm, as shown in the highly magnified I–V characteristic in Fig. 5(b), the resistance was much higher than that of the other cases, but the sample also showed approximately an ohmic behavior. Although we have to optimize the device structures and processes to improve the resistance of the hybrid structures in the current work, the resistance decreased with decreasing the distance between the NWs. The sample also showed a rectifying diode property within the current range, i.e., nanoampere, nA, similar to that shown in Fig. 5(b). These results showed that the hybrid NWs had p-type conductivity.

In our previous study, 41) we found the electrical and magnetotransport characteristics of the samples with MnAs NCs formed on GaInAs/InP (111)B layers by conventional MOVPE. The MnAs NCs were self-assembled on GaInAs (111)B surfaces at the growth temperature T₉ of 650 °C, and all the samples showed p-type conductivity with nearly the same carrier concentrations of approximately 1.7 × 10¹⁸ cm⁻³ estimated at 280 K. 41) Therefore, as in the case of the self-assembled formation of MnAs NCs on GaInAs (111)B surfaces, the Mn atoms provided during the endotaxy of MnAs NCs at a T₉ of 600 °C thermally diffuse into the undoped GaAs NWs in the current work. Then, they are unintentionally incorporated as an acceptor yielding p-type GaAs layers, or possibly p-type paramagnetic dilute magnetic semiconductor layers of GaMnAs, near the NW surfaces. We observed relatively large differences in the electrical currents measured for the present hybrid structures between the p- and n-type substrates, i.e., microampere, μA, in Fig. 5(a) and milliampere, mA, in Fig. 6(a), possibly
owing to the poor reproducibility in the fabrication processes of the electrodes and/or the differences in carrier concentrations near the NW surfaces. However, we believe that the electrical characteristics of hybrid structures between ferromagnetic NCs and semiconducting NWs are much improved by introducing InAs NWs, which have been demonstrated in our vertical surrounding gate transistors, as an electrical channel of core NWs, in combination with the optimization of the device processes. Further detailed growth experiments are required for controlling the endotaxial nanoclustering of MnAs in InAs NWs.

4. Conclusions

We fabricated MnAs/GaAs hybrid NWs by combining SA-MOVPE of undoped GaAs NWs and endotaxial nanoclustering of MnAs. As a result of endotaxy, MnAs NCs were typically embedded in the six ridges of hexagonal GaAs NWs. However, the MnAs NCs were formed not only at the six ridges, but also on the surfaces of six facets of the NW templates, six ridges, but also on the surfaces of six facets of the NWs. However, the MnAs NCs were formed not only at the bottom part of the NWs. To characterize electrical properties of MnAs/GaAs hybrid NWs, two-terminal device processes for free-standing hybrid NWs were developed. Current and voltage characteristics showed that the hybrid NWs formed on Zn-doped p-type GaAs (111)B substrates had ohmic characteristics, while those formed on Si-doped n-type GaAs (111)B substrates had clear rectifying characteristics. The hybrid NWs showed p-type conductivity possibly owing to the formation of p-type GaAs layers near the NW surfaces caused by the thermal diffusion of Mn atoms into GaAs NWs during the endotaxial nanoclustering of MnAs.

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