論文の内容の要旨

論文題目 Spin dependent transport phenomena in III-V semiconductor heterostructures with ferromagnetic MnAs nano-scale particles

（ナノスケール強磁性MnAs微粒子を含むIII-V半導体へテロ構造におけるスピン依存伝導現象）

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In semiconductor-based spintronics devices, novel functionalities are expected by utilizing not only the charge but also spin of carriers. The realization of such devices relies both on the ability to inject a spin-polarized current into a semiconductor (SC) and to detect this current. One of the recent approaches is using ferromagnetic semiconductor (FMS) layers as a spin injector or a detector in device structures. However, FMSs have a disadvantage of low Curie temperature (T_C). Another approach is using ferromagnetic metals (FM) as spin injecting and detecting layers. In this approach, however, interfacial compound layers are easily formed, thus the spin polarizations of carries at the interface is dramatically degrade. In 1990's, some magnetic Mn compounds such as MnAs, MnGa and MnAl were successfully grown on semiconductor substrates. Among them, the ferromagnetic metal MnAs is a promising material because its T_C = 313 ~ 318°K and it can be epitaxially grown without interfacial reaction on many semiconductor substrates such as Si(001), Si(111), GaAs(001) and GaAs(111B), although the crystal structure of MnAs (NiAs-type hexagonal) is different from that of GaAs or Si. However, electrical probing spin injection from MnAs into SC still remains a challenging issue because it is not easy to grow MnAs / SC / MnAs heterostructures. Due to the difference of crystal structure between MnAs and SC, overgrowth of SC layers on MnAs is very difficult.

On the other hand, the GaAs:MnAs granular material which contains ferromagnetic MnAs nanoparticles embedded in a GaAs matrix has the advantage of good compatibility with III-V heterostructures and room-temperature ferromagnetism. The GaAs:MnAs granular material is formed by phase decomposition and phase separation in meta-stable GaMnAs alloy semiconductor annealed at 500-700°C . When annealed at around 500°C, zinc-blende (ZB) MnAs nanoparticles, which correspond to nano-scale areas with very high Mn concentration, are formed due to phase decomposition of GaMnAs. When annealed at higher temperature (> 550°C), the more stable hexagonal NiAs phase precipitates. Although the magneto-optical property of hexagonal or zinc-blende MnAs nanoparticles was intensively investigated, its spin dependent transport is little studied. If spin dependent transport of hexagonal or zinc-blende MnAs nanoparticles is realized, they can be used as spin injecting and spin detecting sources in FM / SM multi layers. Furthermore, because the size of MnAs nanoparticles is as small as 2-10 nm, the Coulomb Blockade (CB) effect is supposed to appear, thus MnAs nanoparticles can be used in single-electron devices. Finally, MnAs nanoparticles can also be used in many important parts of quantum computers using quantum dot systems, such as sources of local magnetic field, spin injectors, and spin detectors of the dot spins. Detailed investigations on the spin dependent transport as well as the charge effect in semiconductor nanostructures containing MnAs nanoparticles will be very important for understanding basic phenomena that can help us to develop new kinds of nano-scale spintronic devices.

This thesis presents studies of spin dependent transport phenomena in semiconductor based heterostructures consisting of MnAs thin film / III-V semiconductor / GaAs:MnAs granular layer, magnetic behaviors of MnAs nanoparticles, and device applications of ferromagnetic nanoparticles in active devices.

First, high quality single crystal magnetic tunnel junctions (MTJs) consisting of MnAs thin film / GaAs / AlAs / GaAs: hexagonal MnAs nanoparticles were grown by molecular beam epitaxy. TMR of those MTJs was clearly observed for the first time. We show that by using hexagonal MnAs nanoparticles as ferromagnetic electrodes, semiconductor based MTJs with high TMR ratio (18 % at 7 K), high V_{half} (1200 mV at 7 K) and high operating temperature (~ 300 K), were obtained. The observed TMR ratio is the highest among MnAs-based MTJs. The TMR ratios were found to oscillate against the tunnel barrier thickness while the tunneling resistances follow the WKB approximation, revealing that there are some kinds of quantum effects in our single crystal MTJs. This result shows that hexagonal MnAs nanoparticles can be used as spin injecting and spin detecting sources in semiconductor based spintronics devices.

Next, magnetic properties of hexagonal MnAs nanoparticles were studied by spin dependent transport measurements and Monte Carlo simulations. By utilizing the TMR effect in MnAs thin film / GaAs / AlAs / GaAs: hexagonal MnAs heterostructures, we observed rich magnetic behaviors of hexagonal MnAs nanoparticles system, such as their static and dynamic *M-H* characteristics, relaxation of magnetization, and blocking temperatures. By fitting the Monte Carlo simulations to the *M-H* data at 7 K, we estimated the anisotropy constant of hexagonal MnAs nanoparticles to be about 2.1×10^5 ~ 2.5×10^5 ergs/cc. Furthermore, hexagonal MnAs nanoparticles are found to have long relaxation times (several hundreds ms at 7 K for particles with ϕ = 5nm) as well as high blocking temperature (T_B = 230 K for particles with ϕ = 5nm, and T_B ~ 300 K for particles with ϕ = 10 nm). Those magnetic behaviors cannot be explained by Neel model for non-interacting particles. We suggested that the dipolar interaction is the origin of such long relaxation time and high blocking temperature.

Then, we studied the CB effect of double barrier MTJs consisting of hexagonal MnAs nanoparticles. The charging energy and the capacitance of hexagonal MnAs nanoparticle with ϕ = 5 nm are estimated by measuring the temperature dependence of conductance of GaAs:MnAs granular layers. The measured values are consistent with theoretical values and show dependence on the surrounding environment. The TMR oscillation of hexagonal MnAs based double barrier MTJs due to CB was observed for the first time both in vertical and lateral structures. The TMR oscillation curve observed in the lateral structure reveals that the spin-relaxation time of MnAs nanoparticles is as long as 10 μs.

We then propose a new type of spin device called single-electron spin transistor (SEST) that utilizes the TMR and CB effects. We show that by using SEST, reconfigurable circuits such as Tucker type inverter, AND/OR reconfigurable gate, reconfigurable logic gates for two input all symmetric Boolean functions and for two input asymmetric Boolean functions can be realized without using any floating gates. The proposed logic gates can provide nonvolatile and scalable reconfigurable hardware for future electronics.

Finally, electromotive force (emf), Coulomb blockade, and huge magnetoresistance in MTJ with zinc-blende MnAs nanoparticles are described. By detailed theoretical and experimental investigations, we show that the observed emf results from the conversion of the magnetic energy of ZB MnAs nano-magnets into electrical energy when these nano-magnets undergo magnetic quantum tunneling. Our results strongly suggest that Faraday's Law of induction must be generalized in order to account for purely spin effects in such magnetic nanostructures.

Although the subjects of this study are limited to nanoparticles of manganese arsenide compounds, our results provide the basic ideas and experimental foundations for any future studies of spin dependent transport phenomena in ferromagnetic semiconductor granular materials. Recently, during the quest for ferromagnetic semiconductors worldwide, many families of nanoparticles have been discovered. Most of them show room-temperature ferromagnetism. Thus, it is our hope that ferromagnetic nanoparticles embedded in semiconductor materials will be extensively studied and find their application in novel spin devices in near future.

The main achievements of this research are as follows: we experimentally observed the TMR and CB effects, as well as their related phenomena in III-V based MTJs containing MnAs nanoparticles, to better understand magnetic behaviors of MnAs nanoparticles, and to design new kind of devices utilizing the TMR as well as CB effects. A new spin effect, namely the electromotive force induced by a static magnetic field, has been discovered in MTJs with zinc-blende MnAs nanoparticles.