博士論文(要約)

Properties and device applications of new Fe-based ferromagnetic semiconductors and heterostructures

(Fe ベース新規強磁性半導体および そのヘテロ構造の物性とデバイス応用)

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Spintronics is the field of science and technology that aims to utilize both the charge and spin degree of freedom of electrons in device applications. This combination is expected to create a new generation of devices with low-power consumption and new functionalities, which lead to an evolutionary change in the way we are storing, analyzing, and transferring information. One central challenge of spintronics is to integrate spin-dependent phenomena into semiconductors, the platform of almost all present electronic devices. So far, the most straightforward way is to create ferromagnetic semiconductors (FMSs), which is usually done by doping a large amount of transition metal elements (Mn, Fe, Cr…) into conventional semiconductors.

Researches on FMSs so far have been concentrated on Mn-based III-V FMSs like (Ga,Mn)As or (In,Mn)As. However, the availability of only p-type FMSs, the low Curie temperatures (T_C), and the complicated band structure in these materials have hindered their device applications. While many efforts in searching for an ideal FMS have been concentrated on wide-gap diluted magnetic semiconductors such as oxides, nitrides, carbides, our early works on Fe-based III-V FMS (In,Fe)As have shown many positive results. (In,Fe)As was proved to be the first n-type electron-induced III-V FMS with strong *s-d* exchange interaction. These results indicate that the narrow-gap FMSs also possess a very high potential.

This thesis presents the magnetic properties and device applications of new Fe-based ferromagnetic semiconductors (FMSs) and their heterostructures. A large part of the study focused on the n-type FMS (In,Fe)As. The magnetic properties of (In,Fe)As were studied in ultrathin heterostructures, in field-effect-transistors, and in spin Esaki diodes. The results obtained in these works shed light on many fundamental characteristics of (In,Fe)As: The exchange interactions between the electron carriers and the localized Fe magnetic moments, and the band structure. This insightful knowledge enabled us to discuss on the mechanism of ferromagnetism of (In,Fe)As, and propose a chemical trend of Fe-based FMSs. Finally, studies on another Fe-based FMSs such as (Al,Fe)Sb was also conducted and the properties of (Al,Fe)Sb are presented.

In chapter 2, the magnetic properties of ultrathin InAs/(In,Fe)As/InAs trilayer structures grown on GaAs were studied. The quantum size effect was observed in these ultrathin heterostructures using magnetic circular dichroism (MCD) spectroscopy. The results indicate that electron carriers of (In,Fe)As possess a long coherence length (>40 nm) and the their wavefunctions extend into the neighboring InAs layers. This is the manifestation of the *s*-orbital nature of the electron carriers in (In,Fe)As. Using wet etching method to gradually reduce the thickness of the top InAs layer of a 5 nm InAs/5 nm (In,Fe)As/5 nm InAs trilayer quantum well (QW), we observed a large decrease of T_C by 55% without modifying the (In,Fe)As magnetic layer. These results were successfully explained by the decrease of the overlap of the electron wavefunctions and the (In,Fe)As magnetic layer in the QW with etching (the *wavefunction engineering of ferromagnetism*). A large *s*-*d* exchange interaction energy $N_0\alpha$ value of 4.5 eV was estimated for (In,Fe)As QWs based on the mean-field Zener model for two-dimensional magnetic system. This value is highly unexpected and requires reconsideration of the theoretical understanding of the *s*-*d* exchange interaction and the mean-field Zener model.

In chapter 3, using the wavefunction engineering of ferromagnetism method, we demonstrated the electrical control of the magnetic properties of InAs/(In,Fe)As/InAs trilayer quantum wells (QWs) in field-effect transistors. Two devices (A and B) with different QW structures were prepared: The QW in device A consists of InAs (2nm)/($In_{0.94}$,$Fe_{0.06}$)As (8nm)/InAs (5nm) on AlSb (50nm), and the QW of device B consists of InAs (2nm)/($In_{0.94}$,$Fe_{0.06}$)As (5nm)/InAs (2nm)/InAs:Si (5nm, Si 5×10^{18} cm^{-3}) on AlSb (300nm). By applying a gate voltage V_G to manipulate the overlap of the two-dimensional wavefunctions and the (In,Fe)As layer, we were able to decrease the T_C in device A from 24 K to 14 K, while increase T_C in device B from 26 K to 35 K. The ability of customizing the T_C - V_G relationship by appropriate QW structure design is an advantage in device applications. Unlike in the conventional experiments, by controlling the carrier wavefunction instead of the carrier density, we also showed that the power consumption can be reduced to only 10^{-4} - 10^{-6} of that of the conventional method. This result provides a new approach for versatile, low power, and ultrafast manipulation of magnetization.

In chapter 4 we studied the band structure of n-type ferromagnetic semiconductor (In,Fe)As, using tunneling spectroscopy in n-(In,Fe)As/p-InAs spin Esaki diodes. (In,Fe)As samples with Curie temperature (T_C) of 45 - 65 K show spontaneous spin splitting energy of 40 - 50 meV in the conduction band, whose magnitude depends on the Fe concentration, electron density, temperature and external magnetic field. When rotating the magnetization in the film plane, tunneling anisotropic magnetoresistance (TAMR), which is the result of the *s,p-d* exchange interactions and spin-orbit interactions in (In,Fe)As, reveals different symmetries of the (In,Fe)As band structure components. The observation of TAMR indicates a rich environment of magnetic physics in (In,Fe)As.

It was shown that the mean-field Zener model fails to explain the ferromagnetism in (In,Fe)As, which may be due to the complete neglect of the local inhomogeneity and the limitation of the perturbative approach of the model. Finally, the chemical trend of Fe-based FMSs was discussed, which suggests that the relative position of the Fe-related IB to the CB or VB of the host materials is important for the realization of ferromagnetism in Fe-based FMSs.

In Chapter 5, we describe the growth and characterization of another Fe-based new FMS, the insulating FMS (Al,Fe)Sb. We investigate the crystal structure, transport and magnetic properties of Fe-doped $(Al_{1-x},Fe_x)Sb$ thin films up to $x = 14\%$ grown by molecular beam epitaxy. All the samples show p-type conduction at room temperature and insulating behavior at low temperature. The $(Al_{1-x},Fe_x)Sb$ thin films with $x \leq 10\%$ maintain the zinc blende crystal structure of the host material AlSb. The $(Al_{1-x},Fe_x)Sb$ thin film with $x = 10\%$ shows intrinsic ferromagnetism with a Curie temperature (T_C) of 40 K. In the $(Al_{1-x},Fe_x)Sb$ thin film with $x = 14\%$, a sudden drop of mobility and T_C was observed, which may be due to microscopic phase separation. The observation of ferromagnetism in (Al,Fe)Sb paves the way to realize a spin-filtering tunnel barrier that is compatible with well-established III-V semiconductor devices.

In Chapter 6, we give the conclusions and further outlook of the work on Fe-based FMSs. So far, we have successfully fabricated n-type FMS (In,Fe)As, p-type FMS (Ga,Fe)Sb and the insulating FMS (Al,Fe)Sb. These materials are promising from both the theoretical and practical perspectives. From the theoretical viewpoint, the study on the origin of the unexpected strong *s,p-d* exchange interaction in these materials gave us new understandings on the physics of FMSs, as well as sketched out a new material approach towards high T_C FMSs. Particularly, n-type FMS (In,Fe)As has shown many new intriguing features. The quantum size effect in the (In,Fe)As thin films allows many possibilities of novel quantum device applications. Large spin splitting in (In,Fe)As creates a high carrier spin polarization, which is good for the purpose of spin injection from an (In,Fe)As electrode. Utilizing the quantum size effect and the large spin splitting energy, tunneling magnetoresistance (TMR) effect in the (In,Fe)As double QW structures is expected to be strongly enhanced. (In,Fe)As, (Ga,Fe)Sb and (Al,Fe)Sb all possess a lattice constant around 0.61 nm and thus can be grown on each other with little difficulty. The combination of these Fe-based FMSs are therefore highly promising to create a platform for new spin-related physics and spin devices such as all ferromagnetic p-n junctions, magnetic tunnel junctions, and spin transistors.