論文の内容の要旨

Electronic structures and magnetic properties of Fe-doped ferromagnetic semiconductors
（Fe をドープした強磁性半導体の電子構造と磁性）

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In the emerging field of spintronics, utilizing the spin degree of freedom of materials is thought to be promising to realize new functionalities that conventional electronic devices do not have. One candidate material for such technology is ferromagnetic semiconductors (FMS), which possess the properties of both ferromagnet and semiconductor and have been intensively studied since the discovery of Mn doped III-V semiconductors, (Ga,Mn)As and (In,Mn)As. In these materials, the ferromagnetic (FM) interaction is mediated by carriers, and hence it is possible to manipulate the ferromagnetism by controlling the number of carriers. In fact, it was demonstrated that it is possible to control the ferromagnetism by applying gate voltage or impinging light onto the materials. However, there has been problems for practical applications: the Curie temperatures (T_C) of these materials are still below room temperature, and only *p*-type materials are available.

Recently, Fe-doped ferromagnetic semiconductors, namely, (In,Fe)As:Be, (Ga,Fe)Sb, (Al,Fe)Sb, (In,Fe)Sb, and Ge:Fe, have been synthesized, and have the potential to overcome the above-mentioned problems. First, the Curie temperatures of (Ga,Fe)Sb and (In,Fe)Sb are higher than room temperature, 340 K for 25% Fe-doped (Ga,Fe)Sb and 335 K for 16% Fe-doped (In,Fe)Sb. Second, various types of transport properties are realized: *p*-type for (Ga,Fe)Sb and Ge:Fe, *n*-type for (In,Fe)As:Be and (In,Fe)Sb, and insulating for (Al,Fe)Sb.

In this thesis, their electronic structures and magnetic properties have been investigated using soft x-ray spectroscopy, namely, x-ray absorption spectroscopy (XAS), x-ray magnetic circular dichroism (XMCD), resonance photoemission spectroscopy (RPES), and soft x-ray angle-resolved photoemission spectroscopy (SX-ARPES). Furthermore, we have performed first-principles supercell calculation to obtain further insight into the electronic structures and the origin of the ferromagnetism.

In Chapter 3, the results of XAS and XMCD measurements to study the magnetization process of (Al,Fe)Sb thin films are presented. From magnetization curves measured by XMCD at various temperatures, it was found that nanoscale ferromagnetic domains of 300-400 μ_B exist even at room temperature well above the T_C of 40 K. This was attributed to non-uniform distribution of Fe atoms on the nanoscale.

Chapter 4 is devoted to RPES, SX-ARPES, and first-principles calculation studies of group IV FMS Fe-doped Ge (Ge:Fe). ARPES spectra showed that the Fermi level is located 0.35 eV above the valence band maximum. From RPES spectra, non-dispersive Fe 3*d* states were found to exist at the Fermi level, which was attributed to majority-spin *p*-*d*(t_2) antibonding states and also to minority-spin Fe 3*d*(*e*) states. It was suggested that the ferromagnetic interaction is mediated by double-exchange interaction within the minority-spin Fe 3*d*(*e*) band.

In Chapter 5, the electronic structure of (Ga,Fe)Sb studied by XAS, XMCD, RPES and first-principles supercell calculation is discussed. From XAS and XMCD, it was suggested that Fe takes the $3d^6\underline{L}$ configuration, where $\underline{L}$ denotes a ligand hole. RPES spectra indicated the itinerant and correlated nature of Fe 3d electrons. The calculated electronic structure was very similar to that of (Ga,Mn)As except that the additional electron of Fe compared to Mn occupies the minority-spin *e* states, which may be the source of the nanoscale ferromagnetic order.

Chapter 6 summarizes the electronic structures of Fe-doped III-V semiconductors obtained by first-principles supercell calculation. The electronic structures can be understood on the basis of the electronic structures of Mn-doped III-V compounds except that the additional electron of Fe compared to Mn occupies either a majority-spin *p*-*d*(t_2) hybridized antibonding state ($t_{a,\uparrow}$) or a minority-spin *e* state ($e_\downarrow$).

The $e_\downarrow$ state is preferentially occupied in the cases of (Ga,Fe)Sb and (In,Fe)Sb, where the valence band or *p* level is located high in energy and, therefore, it is more stable for the additional electron to occupy the $e_\downarrow$ state than to occupy the $t_{a,\uparrow}$ state. It was suggested that the ferromagnetism in Fe-doped III-V FMSs originates from the nanoscale fluctuation of Fe atom distribution as already suggested in the preceding chapters.