論文内容の要旨

論文題目 Interaction between magnetic adatoms and surface two-dimensional electron gas (表面二次元電子ガスと磁性不純物間の相互作用)

氏名 高瀬恵子

Two-dimensional electron gas (2DEG) provides a fascinating playground for low-dimensional physics. Representatives of 2DEG are fabricated at heterostructures of semiconductors, the metal-oxide semiconductors, and thin metal films. Another candidate for producing 2DEG was surfaces of metals or semiconductors. The surface state is created at a few layers of the top of materials, i.e. the interface between the material and vacuum, so that it is *intrinsically* two-dimensional. Moreover, the surface state is so sensitive with extrinsic effects, for example, localization or scattering centers given by adsorption of foreign atoms. Therefore, the surface 2DEG has attracted wide attention of researchers of condensed matter physics and been intensively studied by various techniques, such as scanning tunneling spectroscopy and photoemission spectroscopy.

However, study of the surface state by transport measurements has been still developing because of technical difficulty such as leakage of current through the bulk state or fabricating good contacts to the state in ultra high vacuum (UHV). However, these problems have been addressed by applying a micro-four-point probe method with a narrow probe spacing of ~ 20 μ m on a sample surface, fabricated on a substrate of semiconductor such as silicon. This method enables to electrically decouple the surface state with the bulk state.

By using the micro-four-point probe method, transport properties of various

superstructures have been investigated. Among them, Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface, which was formed by depositing In atoms of 1.2 monatomic layer (ML) thick on a Si(111) substrate, was found to show a metallic character in transport from room temperature to ~ 10 K without any phase transitions. In addition, this surface was reported to have a metallic and parabolic dispersing band by photoemission spectroscopy. Therefore, this surface provides a good system to study the interaction of adatoms with the surface state.

In the present thesis, I focused on studying the interaction between magnetic adatoms and the surface two-dimensional electron state provided by Si(111) $\sqrt{7} \times \sqrt{3}$ -In-rec surface as a prototype of surface dilute magnetic systems. Since the screening in two-dimensional electronic systems is generally weaker than that in three dimensional systems, such interactions may be more significant in the surface state. Concretely, by using the micro-four-point probe method, I conducted surface-sensitive and temperature-dependent resistivity measurements for the $Si(111)\sqrt{7}\times\sqrt{3}$ -In-rec surface with various concentrations of Co adatoms *in situ* in UHV. From applying the appropriate theories for the observed temperature dependences of resistivity, magnetic effects in the presence of disorder were found to play a crucial role. Moreover, the observed behavior has a possibility of indicating a change of the magnetic state of the sample surfaces. In the following, I describe the details.

In the dilute concentration regime of the Co coverage from ~ 0.00025 ML to ~ 0.00125 ML, the resistivity deviation from the pristine Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface was first observed at temperatures lower than 100 K for all the sample surfaces, while the pristine Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface remained metallic from room temperature to ~ 10 K with high residual resistivity ratio, indicating a high defect density. At the lowest concentration of Co adatoms of ~ 0.00025 ML, the deviation in resistivity monotonically increased down to the lowest temperature, as shown in Fig. 1(a). At the slightly higher Co coverage, the deviation in resistivity maximum at a certain temperature $T(\rho_{max})$ was linearly proportional to the Co coverage shown in Fig. 1 (b), which indicates that the observed temperature dependence of resistivity was associated with spins induced by Co adatoms.

To clarify the origin of the observed phenomena, I focused on the interaction of conduction electrons and magnetic impurities in the presence of disorder, such as weak localization, electron-electron interaction, and *s*-*d* interaction (Kondo effect, RKKY interaction, and competition between them). The calculated resistivity by quantum corrections of weak localization and electron-electron interaction in the presence of magnetic impurities were too small to explain the magnitude of the observed resistivity

deviation. In addition, the calculation did not reproduce the observed character of Co coverage dependence of $T(\rho_{max})$. However, a competition between Kondo effect and RKKY interaction can explain the observed phenomena, indicating that it was a dominant mechanism. From fitting for the respective samples, I deduced Kondo temperature T_k of the Co/Si(111) $\sqrt{7} \times \sqrt{3}$ -In-rec surface as ~ 4 K which was intrinsically independent on Co coverages, and typical RKKY temperature $T_{\rm sg}$ (corresponding typical RKKY interaction energy) which depended on the Co coverage. The values indicated that the ground state of the sample with Co coverage of 0.00025 ML preferred Kondo state, in which spins are isolated with negligible mutual interaction, while at higher coverages, RKKY interaction between the spins could not be ignored. Moreover, by considering the enhancement factor of coefficient of Kondo logarithmic resistivity by disorder, I found the *quantitative* agreement between the calculation and the observed deviation of resistivity for the sample with the lowest Co coverage, by using a parameter of the deduced T_K only. With increase of Co coverage, however, the calculations showed larger deviation from the observations in the magnitude. It means that it was necessary for the correction of enhancement factor, including the effect of RKKY interaction in the presence of disorder.

Furthermore, it should be noted that the magnetic states of Co/Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface in the dilute range of Co adatoms. It was reported that at $T_K > T_{sg}$ the ground state was Kondo state, while at $T_K < T_{sg}$ a spin-glass state possibly overcame the Kondo state due to the enhanced RKKY interaction. According to this assumption, the magnetic state experienced a transition from the Kondo state to a spin-glass state by increasing the Co coverage.

In the dense regime of Co coverage from ~0.0125 ML to ~ 0.58 ML, the transport results were different from those in the dilute regime. For the sample of Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface with Co adatoms of 0.58 ML, the temperature dependence of resistivity showed a semiconducting character, which was often observed for granular metal. However, for the samples with Co coverage of ~ 0.0125 ML to ~ 0.2 ML, the resistivity first showed a decrease by cooling, indicating a metallic character. Then, it passed through the resistivity minimum, and then turned to increase by further cooling. All of them shows a resistivity higher than that of the pristine Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec or the dilute Co/Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface at room temperature, indicating an enhanced disorder. In addition, the temperature dependence of resistivity showed a change in the slope, leading to the change of effective mass or Fermi wave vector of the samples. From extrapolation of resistivity at higher temperature, I estimated the resistivity raised by spins of Co adatoms for the respective samples. All of them were larger than the resistivity values expected by weak localization and electron-electron interaction, and fitted well with a logarithmic function of temperature. A two-level model of tunneling

electrons, which was a significant mechanism for strongly disordered systems, was found to be inappropriate to explain the phenomena. In addition, the Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface with adsorption of Ag of ~ 0.05 ML, which showed almost the same resistivity with Si(111) $\sqrt{7}\times\sqrt{3}$ -In-rec surface with Co of ~ 0.03 ML at room temperature, did not exhibit the logarithmic rise in resistivity at lower temperature. This supports that the observed deviated resistivity has a magnetic origin.

The conduction mechanism of the present samples in the dense Co regime was considered to be the same as that of Kondo-like resistivity, which was observed in strongly disordered bulk systems with denser magnetic concentrations than in ordinal dilute magnetic alloys. The normalized resistivity by the resistivity minimum and the temperature at the resistivity minimum showed a characteristic dependence of Co coverages. This possibly indicates that the magnetic state showed a transition from spin-glass to inhomogeneous ferromagnetism.

As described as far, in the present thesis, I have carried out temperature dependence of Co/Si(111) $\sqrt{7} \times \sqrt{3}$ -In-rec with various Co coverages. From analysis by using available theories, I found that *s*-*d* interaction in the presence of disorder was essential to explain the observation in the regime of dilute concentration of Co adatoms. In the dense regime, moreover, I found that magnetic interaction in the presence of strong disorder was also important to explain the observation. Finally, I suggest that I have detected three phase of Co adatoms on a metallic surface 2DEG of Si(111) $\sqrt{7} \times \sqrt{3}$ -In-rec by transport measurements with changing the adatom concentration; a dilute phase in which Kondo effect is dominated, a medium-concentration phase in which RKKY interaction occurs and a spin-glass state is possibly formed, and a dense phase where an inhomogeneous ferromagnetism begins to appear with stronger spin-spin interaction. These findings provide an important example of fundamental physics of dilute magnetism on surfaces and also can have possible applications to spintronics.





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The line on the sample is the guide to the eye. (b) The temperature at the resistivity maximum versus Co coverages.