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Large Negative Magnetoresistance of Intercalation Compound $Mn_x TiS_2$

Masasi Inoue [a], Hiroshi Negishi [a], Hiroshi Yamada [a], Saiko Negishi [a] & Minoru Sasaki [a]

[a] Department of Materials Science, Faculty of Science, Hiroshima University, Higashi-Hiroshima, 739, Japan

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Large Negative Magnetoresistance of Intercalation Compound Mn_xTiS_2

MASASI INOUE, HIROSHI NEGISHI, HIROSHI YAMADA, SAIKO NEGISHI, and MINORU SASAKI
Department of Materials Science, Faculty of Science, Hiroshima University, Higashi-Hiroshima 739, Japan

A relatively large negative magnetoresistance has been evidenced in intercalation compounds Mn_xTiS_2 ($0 < x \leq 0.25$) from measurements up to 31 T at 1.8, 4.2, and 77 K. For low Mn concentration $x \leq 0.03$, Mn 3*d* electrons are regarded as 'localized' spins with thermal fluctuation, while for higher concentration $x > 0.03$, a 'Mn-derived band' is formed near the Fermi energy. In the layered structure of Mn_xTiS_2, the carrier transport takes place along the host TiS_2 layers and through the interlayer spaces via intercalated Mn atoms. The observed negative magnetoresistance can be reasonably explained by existing spin fluctuation theories.

Keywords: intercalation compounds; large negative magnetoresistance

INTRODUCTION

Layered transition-metal intercalation compounds M_xTiS_2 are formed by intercalation of 'guest' 3d metals, M, into the van der Waals gaps between the 'host' TiS_2 layers. Depending on the nature of the intercalated species and its concen-

tration, M_xTiS_2 compounds exhibit interesting electronic and magnetic properties[1-3] which can be understood in terms of an 'itinerant electron' model[4-6] rather than a 'localized' picture. In particular, Mn_xTiS_2 shows a paramagnetic behavior down to 4.2 K over a wide x range ($x \leq 0.25$) and a negative magnetoresistance, while the host TiS_2 has a small positive magnetoresistance[7]. However, much less is known about the mechanism of negative magnetoresistance. In the present work, we have studied the transverse magnetoresistance of the paramagnetic Mn_xTiS_2 ($x \leq 0.25$). Using all of the experimental data on this material, we shall discuss its characteristic properties and the mechanism of negative magnetoresistance.

EXPERIMENT

Single crystals of Mn_xTiS_2 ($x \leq 0.25$) were grown by a chemical vapor transport technique[8]. Electrical leads with six-probe contacts to a single crystal were made by soldering with pure indium metal. The magnetoresistance was studied up to 31 T at 1.8, 4.2, and 77 K by a conventional dc potentiometric method using a compact home-made pulsed magnet, the magnetic field being applied along the c axis and the sample current being measured along the a axis.

RESULTS AND DISCUSSIONS

The temperature dependence of the electrical resistivity of intercalation compounds M_xTiS_2 (M=3d metals) shows generally a metallic behavior, which can be explained by temperature-independent impurity, and temperature-dependent intra- and inter-valley scatterings[9]. The magnetoresistance $\Delta\rho/\rho_0$ {$= [\rho(H) - \rho_0]/\rho_0$ with $\rho(H)$ at magnetic field H and ρ_0 at $H = 0$} for the host TiS_2 is positive and very small even at liquid helium temperature, of the order of 5% at 31 T; its magnetic field dependence obeys the standard quadratic magnetoresistance law $\Delta\rho/\rho_0 \propto H^2$. On the other hand, for paramagnetic Mn_xTiS_2 ($x \leq 0.25$) crystals, a relatively large negative magnetoresistance $\Delta\rho/\rho_0$ is found.

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The magnetic-field dependence is expressed by the power law, $-\Delta\rho/\rho_0 \propto H^m$, with $m \cong 2$ at 77 K for all samples up to 31 T. At 4.2 K this is not the case, as shown in Fig. 1, which plots the data on a logarithmic field scale. Here the solid lines show the calculated curves over the entire magnetic field range using the spin fluctuation theory (see later). Lightly intercalated crystals with $x < 0.10$ exhibit a saturation at higher field but heavily intercalated ones with $x > 0.15$ do not. At low fields the experimental data follow the power law $-\Delta\rho/\rho_0 \propto H^m$ with the x-dependent exponent m, as given in the inset. The behaviors at 1.8 K are almost the same as those at 4.2 K.

Furthermore, it was found that various properties of $Mn_x TiS_2$ depend on the Mn concentration.[1] Two regions can be distinguished: region I for $x \leq 0.03$, where Mn $3d$ electrons are regarded as 'localized', and region II for $x > 0.03$, where they are spread in a 'Mn-derived band'. In the low Mn concentration range, isolated Mn spins on the intercalated Mn^{2+} ions, that are formed by

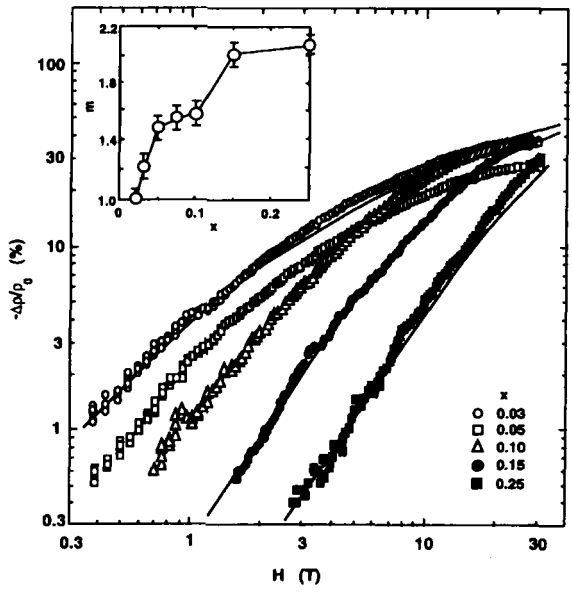

FIGURE 1 $-\Delta\rho/\rho_0$ vs. H at 4.2 K for Mn_xTiS_2. In weak field each curve follows the power law, $-\Delta\rho/\rho_0 \propto H^m$; the exponent m is shown in the inset. The solid lines are the calculated curves according to spin fluctuation theories.

a charge transfer to the TiS_2 conduction band (whose energy may be located somewhere above the Fermi energy E_F), are thermally fluctuating in zero field. Upon application of an external magnetic field H, these spins align along H, which suppresses the thermal fluctuation of the localized spins, leading to a reduction in magnetic scatterings for conduction electrons and thus to a negative magnetoresistance. On the other hand, in the high Mn concentration region, a Mn 3*d* derived band is formed near E_F, which, upon application of a magnetic field, is split into up- and down-spin bands. The band nature in intercalation compounds M_xTiS_2 (M = Fe, Ni, and Co) has been predicted theoretically by Motizuki *et al.*[4-6] Hence we believe that the presence of a strong 'hybridization' among the guest and host atoms and thus the nature of the Mn-derived band are the key point for the origin of the negative magnetoresistance observed even in the paramagnetic Mn_xTiS_2 in its high Mn concentration range.

We have proposed a tentative model for the transport process in the layered structure of host TiS_2 and Mn_xTiS_2 in the two Mn concentration regions. It should be first noted that the *c*-axis resistivity ρ_c of the host TiS_2 (perpendicular to the layers) is about 10^2 times larger than the *a*-axis resistivity ρ_a (parallel to the layers)[9], which means that this material has two-dimensional character with a main conduction path along the *a*-axis (*intralayer conduction*). As described earlier, self-intercalated Ti atoms exist in the form of Ti^{4+} or Ti^{3+} in the van der Waals gaps[10]. Then for TiS_2 we assume that only the *a*-axis conduction dominates [Fig. 2(a)]. Since TiS_2 shows only a small positive magnetoresistance, the intralayer conduction is here assumed to be magnetic-field independent. For the intercalate Mn_xTiS_2, on the other hand, the ratio ρ_c/ρ_a is reduced and *interlayer conduction* along the *c*-axis becomes also effective through the intercalated Mn atoms, as depicted in Fig. 2(b). In this case, the conduction path is formed via the Mn atoms and thus conduction electrons pass through a three-dimensional network. The presence of an intercalated Mn atom locally results in formation of a unit cell with the Mn_1TiS_2 formula.

Now the negative magnetoresistance can be understood by existing spin fluctuation theories developed by Ueda[11] applicable for weakly ferromagnetic (WF) and nearly ferromagnetic (NF) metals. Using Ueda's theoretical equations, we have performed computer simulations for the magnetic-field depend-

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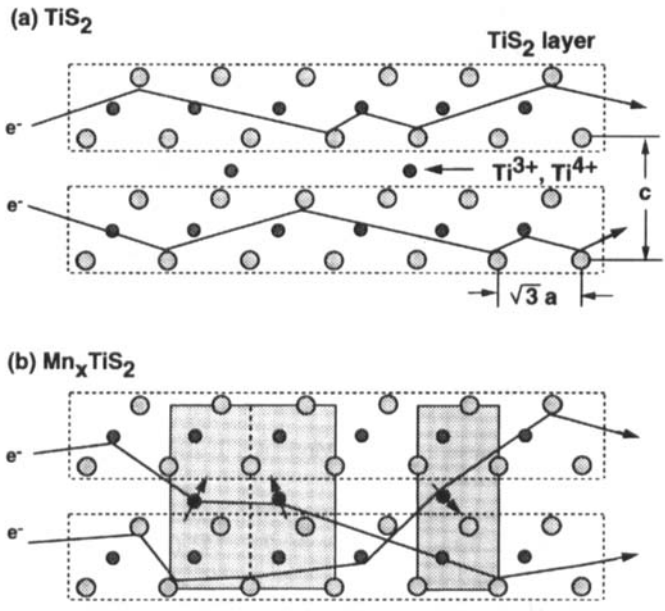

FIGURE 2 Schematic model in the $(11\bar{2}0)$ plane of the crystal for carrier transport (a) along the layers of host TiS_2, (b) through the interlayer space via Mn atoms of Mn_xTiS_2. Shaded regions indicate an isolated Mn-occupied cell or coupled cells.

ence of $\Delta\rho/\rho_0$ over the entire magnetic field range studied, as shown by solid curves in Fig. 1; the curve for $x = 0.03$ ($m = 1$) was calculated using the WF model and those for $x = 0.20$ and 0.25 ($m = 2$) with the NF one. We see that the experimental results are in good agreement with the theoretical curves, except for slight deviation at higher field range for $x = 0.25$. However, the existence of WF and NF orderings has not yet been clearly observed in the magnetic measurements[12,13]. We found only a slight break around 17 K in the temperature dependence of ac magnetic susceptibility parallel to the plane of the crystal, and the specific heat measurements[14] suggests some magnetic phase transition above 14 K.

More theoretical study will be required to cover the intermediate region with the exponent m ($1 < m < 2$) in the power law of the magnetoresistance, as

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well as experimental works for other guest 3d metals in the intercalation compounds M_xTiS_2 at high magnetic fields.

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