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Cross-sectional STM Study of Impurity States in Diluted Magnetic Semiconductor (Zn,Cr)Te

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Abstract. We performed cross-sectional scanning tunneling microscopy / spectroscopy (STM/STS) measurement on multi-layered structure ($Zn_{0.97}$,$Cr_{0.03}$)Te / buffer undoped ZnTe / substrate p-ZnTe. The result of STS measurements on (Zn,Cr)Te clearly showed the existence of Cr impurity states at a deep position within the band-gap of the host ZnTe.

Keywords: Diluted magnetic semiconductor, Scanning tunneling microscopy.
PACS: 75.50.Pp, 81.05.Dz, 81.15.Hi, 68.37.Og, 68.37.Ef

INTRODUCTION

In recent years, so-called "spintronics", which utilize both of charge and spin of carriers, is extensively investigated. Diluted magnetic semiconductors (DMS's) have been energetically studied because of their possibility of application to future semiconductor-spintronics devices. Especially, (Zn,Cr)Te has been regarded as one of the promising materials with its intrinsic room-temperature ferromagnetism when Cr composition is about 20 % [1]. The ferromagnetic interaction between magnetic moments of Cr, which substitute for cation sites in host semiconductor ZnTe is considered to be of short-range character[2], differently from the case of carrier-mediated-ferromagnetism in Mn-doped III-V DMS's such as (Ga,Mn)As, but the mechanism of the interaction has not been clarified. The information on the electronic state of Cr in an atomic scale, such as the spatial distribution of electron orbitals or the local density of states (LDOS), is considered to be essential for the understanding of the Cr interaction, but it has not yet been explored experimentally.

In this study, we performed scanning tunneling microscopy and spectroscopy (STM/STS) measurement for (Zn,Cr)Te, since STM has very high spatial resolution (< 1 Å) in real space, and tunneling current (I_t) of STM is very sensitive to LDOS at a particular energy level corresponding to the applied sample bias voltage (V_s)[3]. Our STM/STS measurement confirmed the existence of the Cr impurity states at a deep position within the band-gap of the host ZnTe.

EXPERIMENTAL METHODS

A multi-layered structure sample was fabricated by molecular beam epitaxy (MBE); a buffer undoped ZnTe (UD-ZnTe) layer (thickness ~100 nm) and a ($Zn_{0.97}$,$Cr_{0.03}$)Te layer (~360 nm) were successively grown on a p-doped ZnTe (001) substrate (P-doped, $1x10^{18}cm^{-3}$) at 573 K. During the growth, the surface was monitored *in situ* by reflection high energy electron diffraction (RHEED). In the STM measurement, a clean cross-sectional (110) surface was obtained by cleaving the sample in a load-lock chamber (~10^{-5} Pa), and then the sample was transferred into an STM chamber of an ultra-high vacuum (< 10^{-8} Pa) without any exposure to air. All STM/STS measurements were performed at 77 K using an electrochemically sharpened polycrystalline tungsten tip (ϕ= 0.3 mm). STM images were obtained with constant current mode, and STS curves were measured in open feedback loop conditions.

RESULTS AND DISCUSSION

Figure 1 (a) shows a typical STM topographic image of the (Zn,Cr)Te/UD-ZnTe/p-ZnTe cross-sectional surface. With an almost homogeneous topographic image, we can hardly see any difference among the three layers. However, we see clear differences in the STS results. Figure 1 (b) shows a mapping image of current-imaging tunneling spectroscopy (CITS) at V_s = +6.5 and -6.5V of the same area as the topographic image (Fig. 1(a)), constructed from tunneling current - sample bias (I_t - V_s) curves measured at 64 x 32 grid points in this area. Distinct features of the respective layers appear in the CITS; the magnitude of tunneling current in the p-ZnTe substrate is large at V_s = ± 6.5 V, reflecting a conductive character of this region, while the tunneling current in the UD-ZnTe and (Zn,Cr)Te regions is much smaller at V_s = -6.5 V, reflecting a semiconductive character of these regions. On the other side, at the positive bias of V_s = +6.5 V, the tunnel-

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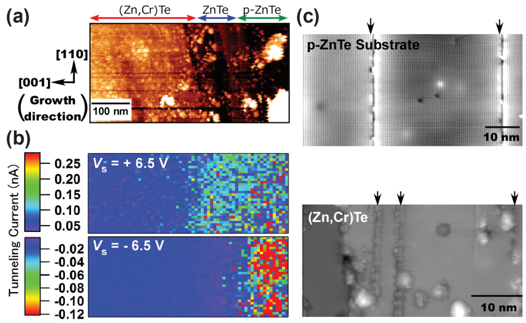

FIGURE 1. (a) STM topographic image of (Zn,Cr)Te / ZnTe / p-ZnTe multi-layered structure (V_s = +6.0 V, I_t = 40 pA) . (b) CITS measured on the same region as (a) at V_s = +6.5 V (above) and -6.5 V (below) (setpoint: V_s = +6.0 V, I_t = 40 pA). (c) Magnified STM images on the p-ZnTe (above) and (Zn,Cr)Te (below) regions. Arrows indicate the position of line defects due to missing of single atomic rows.

ing current in the (Zn,Cr)Te region is smaller than those in the other two regions. These distinct features of the CITS allow us to indentify the position of the border between the regions, as indicated by arrows on top of Fig. 1 (a).

In order to observe the atomic arrangement on the surface of the respective layers, we performed STM observation in a magnified scale. Figure 1 (c) shows typical magnified topographic images in the p-ZnTe substrate (above) and in the (Zn,Cr)Te layer (below), respectively. In the (Zn,Cr)Te region, we did not see any precipitates of a different crystal structure from the zinc-blende structure, except for small amount of adsorbate clusters. Furthermore, several line defects appear along the [110] direction also in the (Zn,Cr)Te region, as indicated by arrows in these images. These line defects are formed due to monoatomic-wide vacancy rows, which are known as intrinsic defects on a cleaved ZnTe(110) surface[4]. The appearance of these line defects in the (Zn,Cr)Te region suggests that the incorporation of a small amount of Cr does not disturb the atomic arrangement on the ZnTe surface.

In order to analyze further the STS character, the I_t - V_s curves averaged over the respective layers are compared, as shown in Fig. 2 (a). As shown in Fig. 2 (a), the I_t - V_s curves in the UD-ZnTe and (Zn,Cr)Te regions have diode-like character; I_t at a negative V_s is much smaller than that at a positive V_s. Since the tunneling current flows mainly through the p-ZnTe substrate to the STM circuit, this kind of diode-like behavior can be explained by the band alignment at the heterojunction of the UD-ZnTe layer and the p-ZnTe substrate; in the present set-up of the STS measurement, a positive (negative) V_s corresponds to applying the forward (reverse) bias. A similar diode-like behavior was also observed in the cross-sectional surface of

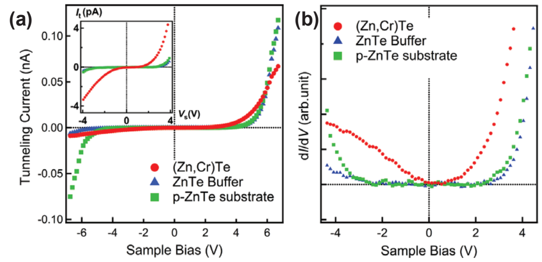

FIGURE 2. (a) STS tunneling current (I_t) - sample bias (V_s) curves measured in the p-ZnTe substrate (green), the buffer undoped ZnTe (blue) and (Zn,Cr)Te (red) regions, respectively (setpoint: V_s = +6.0 V, I_t = 40 pA). The inset shows curves in an enlarged scale between V_s = ±4V. (b) Differential conductance (dI/dV) - sample bias (V_s) curves between V_s = ±4.5V.

undoped AlGaAs/GaAs multiple quantum wells grown on a n^+-GaAs substrate, where a negative V_s corresponds to the forward bias [5].

In order to see the electronic states near the Fermi energy (V_s=0), we focus on the I_t - V_s curves in the range of small V_s. The inset of Fig. 2(a) shows the curves in an enlarged scale between V_s = ± 4 V. In addition, Fig. 2 (b) shows differential conductance vs. bias (dI/dV - V_s) curves, which corresponds to the LDOS at a point of measurement. The dI/dV averaged over the (Zn,Cr)Te region rises at a small value of V_s, in contrast with an almost flat curve in the same V_s range in the UD-ZnTe and p-ZnTe regions. The observed STS feature in the (Zn,Cr)Te region suggests that the existence of an impurity state at a deep level within the band-gap of the host ZnTe. This result is seemingly consistent with an earlier theoretical study [2] which predicted that Cr 3d electrons form an impurity state in the middle of the band-gap of ZnTe and cause ferromagnetic interaction between Cr through the double-exchange mechanism. In this study, we experimentally confirmed the existence of the impurity state by directly observing the electronic state on the surface of (Zn,Cr)Te using STM/STS. We expect further detailed analysis of this impurity state using atomic-resolution STM/STS would contribute toward clarifying the mechanism of ferromagnetism in this material.

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