Spontaneous splitting of ferromagnetic (Ga, Mn)As valence band observed by resonant tunneling spectroscopy

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Current–voltage characteristics of AlAs/GaAs/AlAs double barrier resonant tunneling diodes with ferromagnetic *p*-type (Ga, Mn)As on one side and *p*-type GaAs on the other have been studied. A series of resonant peaks have been observed in both polarities, i.e., injecting holes from *p*-type GaAs and from (Ga, Mn)As. When holes are injected from the (Ga, Mn)As side, spontaneous resonant peak splitting has been observed below the ferromagnetic transition temperature of (Ga, Mn)As without magnetic field. The temperature dependence of the splitting is explained by the the spontaneous spin splitting in the valence band of ferromagnetic (Ga, Mn)As. © *1998 American Institute of Physics.* [S0003-6951(98)03229-X]

Semiconductor resonant tunneling diode (RTD) is a powerful tool for the spectroscopy of the electronic states in the well as well as in the emitter. One can observe spin splitting of electronic states in the paramagnetic/antiferromagnetic ErAs well in the GaAs based AlAs/ErAs/AlAs RTDs under magnetic fields.[1] Local density of states of the heavily doped GaAs emitter were also investigated by a spectroscopy using RTD structures.[2] In order to perform spectroscopy of the valence band structure of *p*-type ferromagnetic (Ga, Mn)As, a III–V based diluted magnetic semiconductor, we have fabricated *p*-type, GaAs based RTD structures using (Ga, Mn)As as an emitter material and studied its current–voltage ($I-V$) characteristics. In this letter we report observation of spontaneous splitting of resonant peaks in $I-V$ characteristics of RTDs below ferromagnetic transition temperature T_C of emitter (Ga, Mn)As, which we interpret as a consequence of valence band spin splitting associated with ferromagnetism in *p*-type (Ga, Mn)As.

We start with a brief description of growth and properties of (Ga, Mn)As. Homogeneous ferromagnetic (Ga, Mn)As has been obtained by molecular beam epitaxy (MBE) at low growth temperatures (200–300 °C), which prevents MnAs formation and Mn surface segregation during growth.[3] Films of (Ga, Mn)As can be grown pseudomorphically on GaAs substrates up to Mn concentration of 7%, above which Mn starts to segregate at the growth front even at low growth temperatures. (Ga, Mn)As based heterostructures, such as superlattice structures[4] and quantum wells,[5] can be realized if one maintains the low growth temperature. A moderate thermal annealing (>600 °C) results in the formation of ferromagnetic MnAs precipitates in the (Ga, Mn)As host,[6] which itself is an interesting but separate subject. The MBE grown (Ga, Mn)As films were all *p* type with hole concentration in the range of high 10^{18}–low 10^{20} cm^{-3}. T_C was found to be as high as 110 K. The ferromagnetism of (Ga, Mn)As is described well by the indirect exchange interaction between magnetic spins mediated by holes (the Ruderman–Kittel–Kasuya–Yosida interaction).[7] Details of the epitaxial growth and the properties of (Ga, Mn)As were presented elsewhere.[3,7,8]

The RTD structures studied in this letter were grown by MBE and consist of, from the surface side, 150-nm $(Ga_{0.965}Mn_{0.035})As$/with or without 15 nm undoped GaAs spacer/5-nm undoped AlAs barrier/5-nm undoped GaAs quantum well/5-nm undoped AlAs barrier/5-nm undoped GaAs spacer/150-nm Be doped GaAs ($p=5\times10^{17}$ cm^{-3})/150-nm Be doped GaAs ($p=5\times10^{18}$ cm^{-3})/p^+ GaAs substrates. All the layers were grown at 650 °C except for the last (Ga, Mn)As layer grown at 250 °C. A schematic zero-bias valence band diagram of the structures is shown in the inset of Fig. 1(a). The precise band diagram is difficult to draw because of the interplay among the band offset between GaAs and (Ga, Mn)As, the role played by the many-body renormalization due to large hole concentration,[9] and the role of randomness introduced by Mn. From the control experiments, the $(Ga_{0.965}Mn_{0.035})As$ layer studied here is known to show a metallike transport behavior typical of a degenerate carrier system and its T_C is about 70 K. Two kinds of samples with or without a 15 nm GaAs spacer layer on the (Ga, Mn)As side were prepared.

The fabrication process of RTDs starts with 30-μm square mesa formation by conventional photolithography and wet etching. Then, Au/Cr contact metals are evaporated on top of the mesa and a SiO_2 film is formed on top of it. The Au electrode is evaporated on the SiO_2 film, which forms a contact to the Au/Cr electrode via photolithographically defined contact holes in SiO_2. Contact to the backside GaAs:Be layer is formed by In through the *p*-type substrate.

The temperature dependence of $I-V$ characteristic of a sample with a 15-nm spacer layer is shown in Fig. 1(a). Positive bias corresponds to hole injection from the (Ga, Mn)As side. Two kinds of samples, with or without the undoped spacer layer, showed almost the same $I-V$ curves. As temperature is lowered, the resonant tunneling features become sharper. Figure 1(b) shows the numerically differentiated $dI/dV-V$ curves of the curves in Fig. 1(a) to enhance the features. In the negative bias side, where holes are injected into the quantum well from p-GaAs, a series of six

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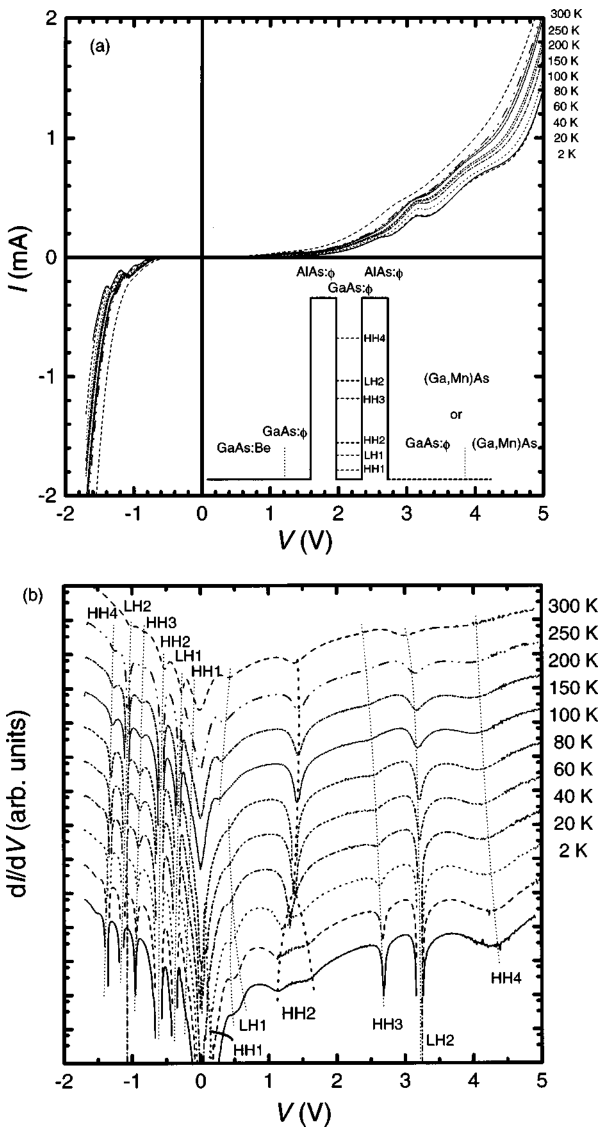

FIG. 1. (a) Current vs voltage ($I-V$) curves and (b) $dI/dV-V$ curves at various temperatures for a RTD with a 15 nm spacer layer. Positive bias corresponds to the hole injection from the (Ga, Mn)As side. The inset of (a) is a schematic valence band diagram of the RTD structures, where hole energy levels are illustrated.

clear resonant peaks with differential negative resistance were observed. These peaks correspond to the six resonant hole levels in the 5-nm GaAs quantum well; from low energy side, heavy hole 1 (HH1), light hole 1 (LH1), HH2, HH3, LH2, and HH4 in order of increasing bias. The number of resonances is in agreement with the previous experiment on *p*-type GaAs RTDs[10] and its theoretical analyses.[11,12] The bias voltage at which the resonance appears shows a linear relationship with the calculated energy levels inside the quantum well[12] as shown in Fig. 2. The slope for the negative bias is 3.0 (Fig. 2) and 2.0 with and without the undoped spacer layer, respectively. This shows the high quality of the present RTD structures, since a slope of 2 is expected for ideal, symmetric RTDs, where the energy of the resonant states in the quantum well is half of the applied voltage at the resonance. In the positive bias side, the six structures were also resolved [Fig. 1(a)], which were somewhat broadened than those in the negative bias side. Broadening of the features is expected since, because of the large number of carriers in (Ga, Mn)As and hence large Fermi energy, one resonance state may start flowing current before the lower energy state turns off. Although the RTD structure is designed to be almost symmetric, asymmetry in $I-V$ characteristics has always been observed. Despite the asymmetry, the linear relationship between the peak voltages and the calculated energy levels is also observed for the positive bias side, as shown in Fig. 2. The slope is 10.0 and 15.5 with and without the spacer layer, respectively. This linear relationship is used later to convert the measured peak position to the energy of the levels. It is not clear at the moment what causes the asymmetry; in actual RTDs, the situation is complicated because voltage drop is expected in electrodes, in depletion region(s) and in accumulation region, in addition to the effect of the series of resistance. The results shown in Figs. 1 and 2, however, clearly demonstrate that resonant tunneling takes place regardless of the emitter material, *p*-GaAs or *p*-(Ga, Mn)As.

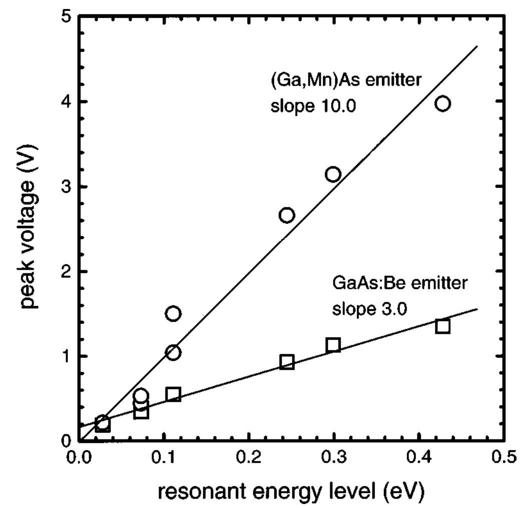

FIG. 2. Peak voltages at 2 K vs hole energy levels inside the quantum well. A linear relationship holds for both positive [(Ga, Mn)As emitter] and negative bias (GaAs emitter).

As seen in Fig. 1(b), the most striking feature is the splitting of the HH2 peak in the positive bias side below 80 K. As temperature is lowered, the HH2 peak becomes sharper down to about 80 K, then it starts to become broader again and at the same time it splits into two peaks. Note that there are no external magnetic fields. No peak other than HH2 showed splitting except for a small trace observed near LH1 of the positive side. Temperature dependence of the splitting in terms of voltage is plotted in Fig. 3. A sharp increase followed by saturation as temperature is reduced bears strong resemblance to the temperature dependence of saturation magnetization of (Ga, Mn)As reported in Ref. 7. In fact, temperature dependence of saturation magnetization calculated from a Brillouin function resulted in an excellent fit shown by the solid line in Fig. 3. Here we assumed $S=5/2$ having Mn^{2+} in mind, although the Brillouin function with other S can similarly fit the temperature dependence. This fit shows that T_C of the present (Ga, Mn)As layer is 72 K, in good agreement with what is expected from control samples. We therefore believe that the splitting observed in the $I-V$ curves is due to the spin splitting of the valence band associated with the development of spontaneous mag-

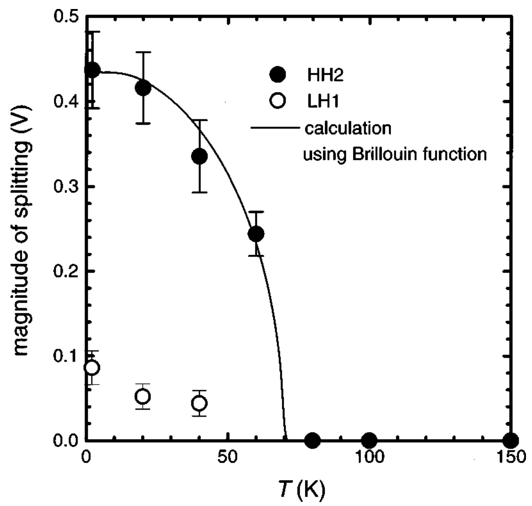

FIG. 3. Spontaneous splitting of peak positions vs temperature. Closed circles are for HH2 peak. A curve based on Brillouin function with $S=5/2$ (solid line) results in an excellent fit. Splitting around LH1 is also plotted in opened circles.

netization in (Ga, Mn)As. The resonant peak LH1 also shows some splitting, which is plotted in Fig. 3 as open circles. The splitting is rather small, and we are not sure whether it is the splitting of the LH1 peak itself or some interference coming from the spin split HH1 peak.

The linear relationship between the voltage and the level energy can be used to obtain the energy splitting ΔE as a function of temperature. At low temperatures it saturates at $\Delta E=44$ meV. We can deduce a measure of the $p-d$ exchange if we simply apply the following expression for the energy splitting in the heavy-hole band:[13]

$$\Delta E = x N_0 \beta \langle S \rangle, \tag{1}$$

where N_0 is the number of cation sites per unit volume, β the $p-d$ exchange, and $\langle S \rangle$ the value of thermal average of magnetic spin. Assuming the full alignment of Mn spins ($S=5/2$) at low temperature, 44 meV splitting gives $N_0\beta=0.6$ eV, much smaller than the one determined from the spin disorder scattering in the transport phenomena at higher temperatures, where $N_0\beta=3.3$ eV.[7] This small value of $N_0\beta$ would lower the transition temperature calculated from the RKKY interaction in Ref. 7 and, to obtain agreement with experiments, one may need to consider additional mechanisms such as ferromagnetic electron–electron interaction due to weak localization, which enhances the ferromagnetic interaction. However, it is perhaps too early to compare the two values determined from the two different types of measurements in the two different temperature regimes. It is not even clear at the moment whether we are observing the heavy-hole band splitting, which gives us Eq. (1). We certainly have to take into account a large mixing between the light and heavy hole states; this would increase $N_0\beta$ at a given ΔE.

The reason why only the HH2 state shows a pronounced splitting is not understood at present. It could be due to the averaging of spin splitting at higher voltage because of a strong band bending at the interface. Or it could be due to some selection rules associated with spin split hole states in the emitter and the spin degenerate resonant states in the well. Further work is needed to answer this question.

In summary, *p*-type GaAs based resonant tunneling diodes with ferromagnetic (Ga, Mn)As as an emitter were prepared by molecular beam epitaxy and their transport properties were studied. When holes were injected from the (Ga, Mn)As side, spontaneous splitting of resonant peaks was observed below the ferromagnetic transition temperature of (Ga, Mn)As without magnetic fields. The temperature dependence of the splitting was proportional to the calculated saturation magnetization, showing that the observed splitting is due to spontaneous valence band spin splitting associated with development of ferromagnetism. This is the first spectroscopy of the spontaneous splitting of valence band by resonant tunneling. The results show the possibility of using a (Ga, Mn)As RTD structure to inject spin polarized carriers into nonmagnetic semiconductors.

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