Observation of spontaneous spin-splitting in the band structure of n-type ferromagnetic semiconductor (In,Fe)As

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The band structure of ferromagnetic semiconductors (FMSs) has been under active debate since their discovery. Ideally, it is expected that the *s,p-d* exchange interactions between carriers and localized magnetic moments would induce spin-splitting in the conduction band (CB) and valence band (VB) of FMSs. If it is the case, we can easily design the material properties by using well-established band engineering of semiconductors. Spontaneous spin-splitting in the VB due to ferromagnetism has been clearly observed only in Mn-based II-VI FMS (Cd,Mn)Te quantum wells (QWs), but the ferromagnetic order disappeared above 4K¹. In Mn-based III-V FMSs such as (Ga,Mn)As, although higher Curie temperature (*T*_C) was obtained, the hole carriers are in the Mn-related impurity band (IB), while the CB and VB of the host materials remain nearly nonmagnetic^{2,3}. The exotic IB picture poses an awkward problem in modeling the material properties and designing devices. FMSs with both large spin-split CB and VB and high *T*_C are thus highly desired for the realization of semiconductor spintronics devices.

Here, we report on the observation of such band structure in n-type FMS (In,Fe)As, using tunneling spectroscopy in (In,Fe)As-based spin Esaki diodes. The device structure, as shown in Fig 1(a), from the surface is 50 nm-thick n-type (In,Fe)As (with or without Be)/5 nm-thick InAs/250 nm-thick InAs:Be (Be concentration 5×10^{18} cm⁻³)/p⁺ type InAs (001) substrate. Two different p⁺n⁺ junction diodes, A and B, were prepared with the (In,Fe)As layers differing in the Fe concentration (6% and 8%, respectively), electron density (by co-doping Be donors at 5×10¹⁹ cm⁻³ in the (In,Fe)As layer in device B), and consequently $T_{\rm C}$ (45 and 65 K, respectively). When the bias voltage V is small, electrons tunnel from the (In Fe)As CB bottom to the p-InAs VB top, thus the tunneling conductance dI/dV directly probes the density of states of the (In,Fe)As CB. Figures 1(b) and (c) show the dI^2/dV^2 -V curves measured at various temperatures in diode devices A and B, respectively. At 3.5 K, the dI^2/dV^2 -V curves show double-valley features, which evolve into single-valley features at temperatures above $T_{\rm C}$ of the (In,Fe)As films. These double-valley features correspond to the spontaneous spin splitting at the (In,Fe)As CB bottom due to the ferromagnetic order at low temperatures. The spin splitting energy ΔE depends on the Fe concentration, temperature (as shown in Fig.1(d)), and external magnetic field. Furthermore, the magnetoresistance of the diodes depends on the direction of the (In,Fe)As magnetization in the film plane, that is, tunneling anisotropic magnetoresistance (TAMR). At different bias voltages, we observed different anisotropy components of the (In,Fe)As band structure distinguished by their TAMR symmetry. The result indicates that the Fe-related IB lies very close to the CB bottom and VB top of (In,Fe)As^{4,5}

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Fig. 1. (a) Device structure and the band diagram (inset) of the Esaki diodes. The (In,Fe)As CB bottom is spin-split. (b)(c) dI^2/dV^2-V curves of devices A and B, respectively, measured at various temperatures (the vertical axes are intentionally shifted for clear vision). Black dots mark the center positions of the valleys obtained by fitting each dI^2/dV^2-V curve by two Lorentzian curves, which correspond to the spontaneous spin splitting at the (In,Fe)As CB bottom (d) Temperature dependence of ΔE in devices A and B.

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