Progress in studied of diluted ferromagnetic semiconductors supported by the REIMI Project: Andreev reflection and electron doping

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Abstract

With the support from JAEA REIMEI project, we have developed novel diluted ferromagnetic semiconductors (DMS) isostructural to iron-based superconductors since 2011. We characterized these DMS systems with magnetic susceptibility, transport, photoemission, and MuSR measurements, and developed theories for electronic states. In this article, we report recent progress on development of a point-contact device, subsequent Andreev reflection measurements, theoretical effort towards n-type DMS systems, and discovery and characterization of an n-type DMS Ba(Zn,Co)2As2.

1. Research Objectives

Diluted ferromagnetic semiconductors (DMS) have been studied extensively since 1990's for potential use in spintronics devices. Prototypical systems, (Ga,Mn)As or (In,Mn)As, however, have limitations as (i) spin and charge co-doping resulting in mostly p-type materials; (ii) poor chemical solubility due to hetero-valence doping of Mn^{2+} to Ga^{3+} site, leading to availability only as MBE films; (iii) ferromagnetic T_C limited to ~ 200 K. Since 2011, with a partial support from the REIMEI funding, part of the present authors developed novel DMS systems, including Li(Zn,Mn)As [1], (Ba,K)$(Zn,Mn)_2As_2$ [2] and $(La_{1-x}Ba_x)(Zn_{1-x}Mn_x)AsO$ [3]. These systems overcome the above-mentioned limitations (i) - (iii), with (i) independent spin and charge doping with p-type carriers, (ii) iso-valent doping allowing synthesis of bulk single crystals, and (iii) highest T_C already reaching 230 K within several years of initial development, with promising

signatures towards room temperature DMS. Furthermore, the "122" DMS systems [2] share the crystal structure with an iron-based superconductor (Ba,K)Fe_2As_2, semiconductor $BaZn_2As_2$ and antiferromagnet $BaMn_2As_2$ with an excellent matching of lattice constants, which would enable making junctions and multilayer heterostructures of various combinations of these ground states. Previously, MuSR studies confirmed bulk ferromagnetism developed in the full volume fraction, with nearly linear relationship between the average static internal field and T_C in p-type systems [1-3]. Photoemission studies confirmed hole-carriers in the top of the valence band [4] and Mn impurity band located well below the valence band maximum [5]. However, synthesis of n-type materials had been unsuccessful, and development of a device was not attempted before 2016, remaining as major challenges for the present Reimei project.

2. Research Contents

(a) Theoretical efforts towards n-type DMS.

Bo Gu and Maekawa performed density functional theory (DFT) and quantum Monte Carlo calculations on electronic structures of DMS system [6], and found that semiconductors BaZn2As2 and BaZn2Sb2, with very narrow energy gap, may become a good host material to support DMS systems with n-type carriers.

(b) Development of an n-type DMS $Ba(Zn,Co)_2As_2$.

In 2017, a part of present authors from Zhejiang University succeeded in synthesizing the first n-type DMS material Ba(Zn,Co)2As2, with ferromagnetic TC up to 50 K [7]. Magnetic moments are doped with Zn2+/Co2+ substitutions while electrons are doped with Zn2+/Co3+ substitutions. The negative sign of charge carriers is confirmed by Hall effect. With the present REIMEI project, we performed MuSR measurements at TRIUMF in 2017.

(c) Andreev reflection from a point contact of $(Ba,K)(Zn,Mn)_2As_2$ and Pb

Single crystals of a p-type DMS $(Ba,K)(Zn,Mn)_2As_2$ was developed at the Institute of Physics in Beijing by a part of the present authors, and a point contact junction with a Pb electrode was fabricated [8], as shown in the inset of Fig. 1. This was the first case of a device based on the novel DMS systems developed by the present research team. The differential conductance G is shown to decrease below the superconducting Tc of Pb, confirming features expected for Andreev Reflection.

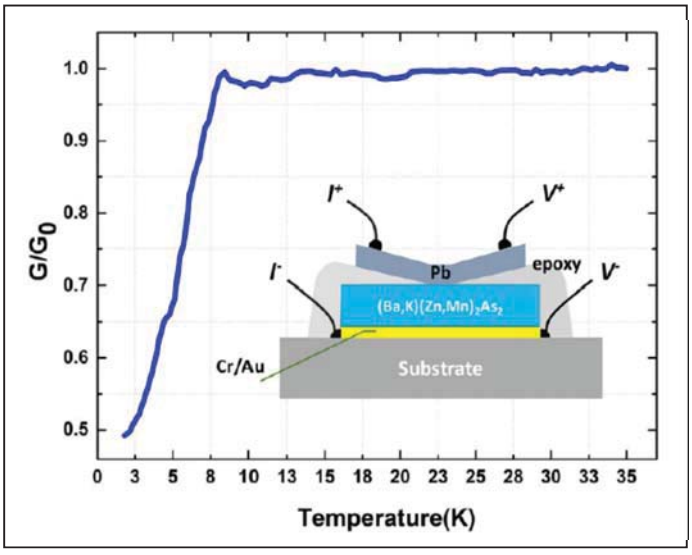

Fig. 1. Normalized differential conductance G/G0 and a sketch for a point contact device between the 122 DMS and Pb used in the Andreev reflection studies [8].

3. Research results

(a) MuSR results on n-type DMS $Ba(Zn,Co)_2As_2$

Zero-field MuSR measurements were performed at TRIUMF on a poly crystal specimen of an n-type DMS system $Ba(Zn_{0.95}Co_{0.05})_2As_2$. Onset of relaxation due to static random local field was observed below the Curie temperature T_C ~ 40 K. In Fig. 2, we plot the internal static field strength at T = 2 K versus T_C observed in the present specimen with earlier MuSR results in other p-type DMS systems [7]. The point for the present n-type system lies at a location very different from the linear trend shown by many other p-type DMS systems [1-3]. Since the static internal field parameter is proportional to the concentration multiplied by the average static moment size in dilute spin systems, the trend for the n-type system implies that T_C is relatively high for a given size and density of the static ordered moments. Hence the ferromagnetic exchange coupling is much larger in the n-type system compared to the p-type systems.

This tendency can be partly ascribed to the difference between the present Co-doped system and Mn-doped p-type 122 DMS systems, which involve frustration because the nearest-neighbor Mn pairs are coupled antiferromagnetically, as can be seen in $BaMn_2As_2$ being a strong antiferromagnet with T_N ~ 625 K. In contrast, $BaCo_2As_2$ is a paramagnet showing a tendency towards ferromagnetic correlation [9]. Therefore, there is no frustration between neighboring Co spins in the Co-doped 122 system. This could lead to the stronger ferromagnetic coupling in the n-type Co-doped 122 DMS as compared to p-type Mn-doped DMS systems. This difference can also explain why the Co-doped 122 DMS system shows a very small coercive field of ~ 20 G, while the p-type Mn doped 122 DMS has a coercive field of ~ 2 T [2].

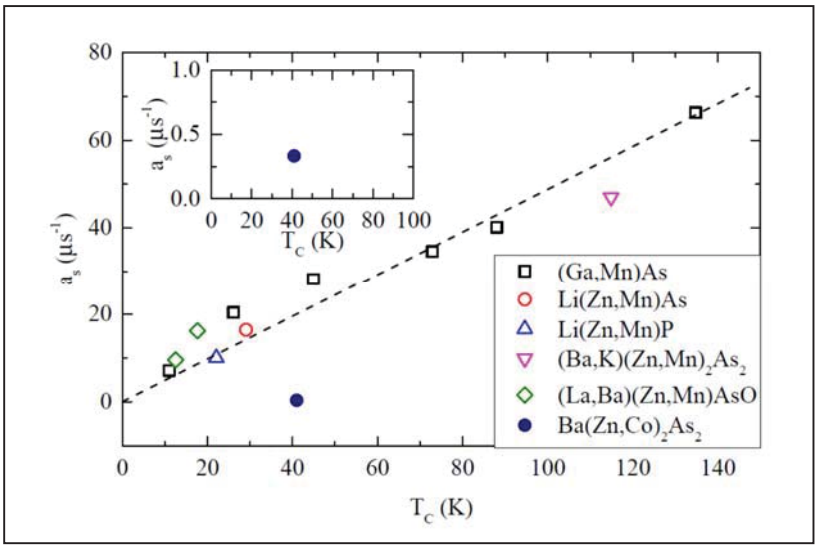

Fig. 2. Correlation between the static internal field parameter as determined at T = 2 K by ZF-μSR versus the ferromagnetic Curie temperature T_C observed in p-type (Ga,Mn)As [10], Li(Zn,Mn)As [1], (La,Ba)(Zn,Mn)AsO [3], $(Ba,K)(Zn,Mn)_2As_2$ [2], and n-type $Ba(Zn,Co)_2As_2$ [7].

(a) Andreev Reflection from a point contact of p-type 122 DMS and Pb

Using a device shown in Fig. 1, the differential conductance was observed as a function of bias voltage and temperature. The conductance depends on the spin polarization of the up-spin band and the down spin band P, which is defined as $P = (N\uparrow - N\downarrow)/(N\uparrow + N\downarrow)$ where $N\uparrow/N\downarrow$ is the density of state for spin up/down band. The results in Fig. 3 can be fit to a model with

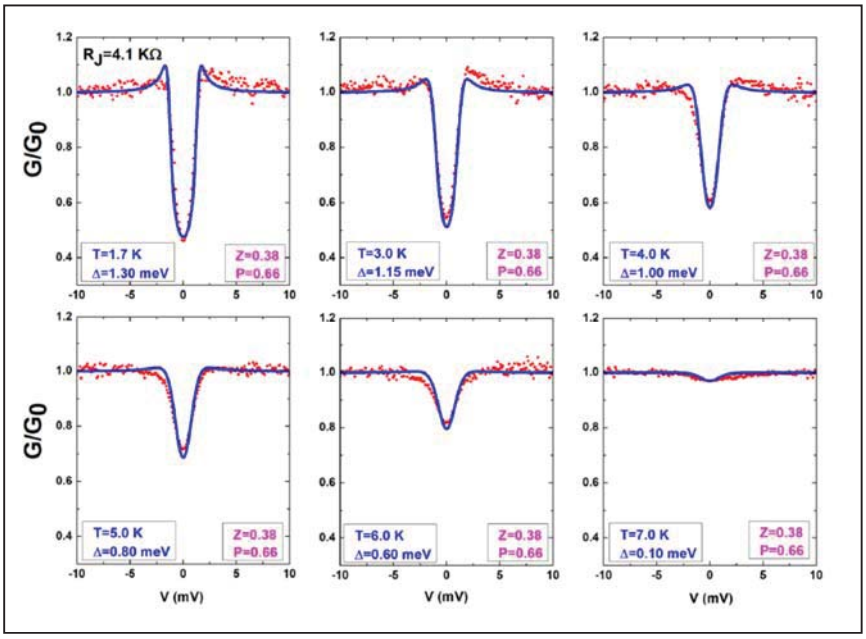

Fig. 3. Normalized differential conductance G/G0 spectra (red dot) from a point-contact device of Pb and $(Ba,K)(Zn,Mn)_2As_2$ with K 0.096 and Mn 0.195 and their fits to the modified BTK theory (blue line) at selected temperatures from 1.7 K to 7.0 K. [8]

the modified Blonder–Tinkham–Klapwijk (BTK) theory, with the spin polarization P of the DMS and the superconducting gap Δ of Pd as parameters. The observed results fits well to P = 0.66, which is consistent with a large spin polarization for a ferromagnetic DMS system [8].

4. Conclusion

We reported progress of the studies of novel DMS systems, in (1) the formation of a point-contact device using a single crystal; (2) successful observation of Andreev Reflection; (3) development of theory describing advantage of a narrow-gap 122 system for possible formation of n-type DMS system; and (4) successful synthesis and characterization of the novel n-type DMS $Ba(Zn,Co)_2As_2$.

5. References

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