

Exchange interactions and microscopic origin of ferromagnetism in dilute magnetic semiconductors

K. Sato^{1,2}, V. A. Dinh³, T. Fukushima^{2,4}, T. Kakeshita¹, and H. Katayama-Yoshida⁵

¹ Graduate School of Engineering, Osaka Univ, Osaka, Japan

² Center for Spintronics Research Network (CSRN), , Osaka Univ, Osaka, Japan

³ Japan International Cooperation Agency (JICA), Japan

⁴ Institute for NanoScience and Design Center, Osaka Univ, Osaka, Japan

⁵ Center for Spintronics Research Network (CSRN), The Univ of Tokyo, Tokyo, Japan

E-mail: ksato@mat.eng.osaka-u.ac.jp

Semiconductors doped with magnetic transition metal impurities are called dilute magnetic semiconductors (DMS) [1]. They are expected to be a key material for realizing semiconductor spintronics due to carrier-induced ferromagnetism observed in these systems. So far, several models have been proposed to investigate the ferromagnetism, and many first-principles calculations have been performed to clarify the relationship between electronic structure and the origin of the ferromagnetism in DMS [1-3].

In this paper, firstly we have performed electronic structure calculations by using the Korringa-Kohn-Rostoker coherent potential approximation (KKR-CPA) method to describe substitutional disorder in DMS [4]. Most of the present calculations were done within the local density approximation (LDA), and in cases we introduced self-interaction correction (SIC) [5] to assess the impropriety of the LDA in the description of the electronic structure. The effective exchange interactions (J_{ij}) between magnetic impurities were estimated by using the Lichtenstein's formula and the interactions were decomposed into each orbital combination [6]. To avoid the complication due to the magnetic percolation effect and focus on the discussion about the mechanism, simple mean-field approximation was employed for calculating Curie temperature. Therefore, in the present T_C estimations we cannot expect reasonable accuracy [2]. In Fig. 1, we plot nearest neighbor exchange interactions between V, Cr, Mn, Fe and Co in GaAs. As previously reported, the interactions are ferromagnetic for V, Cr and Mn, and anti-ferromagnetic for Fe. For Co the interaction is very small. From the decomposition of J_{ij} , we can recognize that the ferromagnetic contribution mainly comes from t_2 - t_2 orbital combination and this corresponds well to the double exchange picture [2]. We can also understand that the negative interaction for Fe is originated from the anti-ferromagnetic super-change interaction between e and t_2 orbitals. This super-exchange interaction changes its

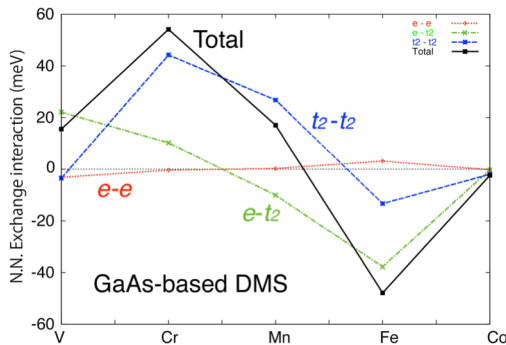


Fig. 1: Calculated nearest neighbor exchange interactions between magnetic impurities in GaAs. The interactions are decomposed into $e-e$, $e-t_2$ and t_2-t_2 contributions. Positive value indicates ferromagnetic interaction.

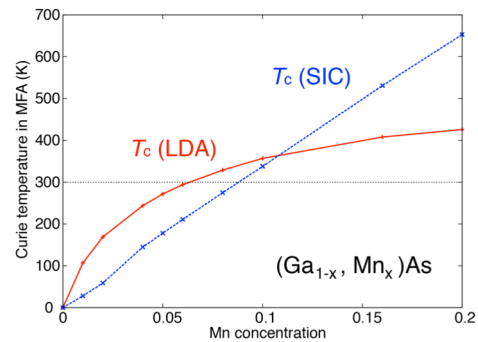


Fig. 2: Concentration dependence of T_C of (Ga, Mn)As in mean-field approximation calculated by the LDA and the SIC.

sign and becomes positive for V. For all cases, $e-e$ contribution is very small due to their non-bonding nature.

So far, our calculations have been based on the LDA result, and the magnetic interaction seems consistent to the double exchange picture. However, it is well known that the LDA systematically overestimate occupied d -states and this might modify the above discussion slightly. For example in Fig. 2, T_C values of (Ga, Mn)As calculated within the LDA and the SIC are plotted as a function of Mn concentration. In the SIC, the concentration dependence becomes more linear and this indicates dominance of the $p-d$ exchange mechanism. Since the anti-ferromagnetic super-exchange is suppressed due to the large exchange splitting in the SIC calculation, calculated T_C becomes large for higher Mn concentration. In summary, the ferromagnetism in DMS is reasonably explained owing to the detailed calculation by the decomposition of exchange interactions.

Thus, ferromagnetism (FM) in GaMnAs has been recognized to be induced by the $p-d$ exchange interaction of the valence holes and localized d -electrons of Mn atoms. Following this model, the d -orbitals of Mn hybridize the p -orbitals of As and form an Mn-induced impurity band (IB) that merges with the valence band (VB) to expand VB towards the bandgap, and the Fermi level (E_F) lies in the merged VB. Besides many experimental works support this model, several experiments have recently found an Mn-induced IB that separated from VB in GaMnAs and E_F may locate in that separated Mn-induced d -band inside the bandgap [7]. The fact that the separation of IB from VB is not found in electronic structure obtained by previous theoretical studies, therefore; in order to explain this finding it is necessary to employ a more reasonable method to deal with this problem.

Lastly, we will show that the above-mentioned finding of the separated IB in GaMnAs can be explained by using the hybrid functional method HSE06 implemented in VASP code [8]. To do that, we first find the most preferable configurations of Mn at various concentrations by a full optimization calculation. We then calculate the band structure and decomposed charge density of the most preferable configuration. We found the tendency for forming the Mn clusters in the host material GaAs and the change in band structure of GaMnAs with respect to the Mn concentration in Mn clusters. Our results show an overlap of IB and VB at the dilute concentrations of Mn atoms in Mn clusters, which is well known as the typical picture of FM originated from the $p-d$ exchange interaction. However, at more condensed concentrations of Mn in Mn clusters, the separation of IB from VB was expanded. This finding suggests that the picture of the origin of FM in GaMnAs would change due to the clustering tendency of the substitution Mn atoms. A comparison of the band structure of GaMnAs and InMnAs corresponding to various Mn clusters configurations will also be given.

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References

- [1] T. Dietl and H. Ohno, Rev. Mod. Phys. **86**, 187 (2014).
- [2] K. Sato et al., Rev. Mod. Phys. **82**, 1633 (2010).
- [3] T. Dietl, K. Sato et al., Rev. Mod. Phys. **87**, 1311 (2015).
- [4] KKR-CPA package (MACHIKANEYAMA2002) H. Akai, <http://kkriissp.u-tokyo.ac.jp/jp/>
- [5] A. Filippetti et al., Phys. Rev. B **84**, 195127 (2011).
- [6] T. O. Kavashnin et al., Phys. Rev. Lett. **116**, 217202 (2016).
- [7] S. Ohya, K. Tanaka and M. Tanaka, Nature Phys. **7**, 342 (2011).
- [8] G. Kresse and J. Furthmüller, Comput. Mat. Sci., 6:15, 1996, Phys. Rev. B, 54:11169, 1996.