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Distance effect on magnetic coupling of Cr doped Bi_2Se_3: A first-principle and Monte Carlo study

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Abstract. In this paper, we investigated the magnetic properties of Cr doped Bi_2Se_3 combining Monte Carlo simulation with first principles method. Our results demonstrate that Cr atoms can be stably doped into Bi_2Se_3. Hysteresis loops were observed in the Cr doped system. The exchange interaction energies of magnetic atoms are calculated at the level of meV. The distance effect is studied with Cr atoms doped in different sites. The magnetic coupling of the atoms inner layer has great impact on magnetic materials, while the interlayer case has an obvious impact on coercivity of the material. However, as the distance of magnetic atoms exceed the quintuple layers (QLs), the magnetic coupling makes hardly contribution to the magnetic system. Our results may further promote the understanding of magnetic coupling in magnetic topological insulator.

1. Introduction

Topological insulator (TI) has become a research hotspot for its special quality that behaves as an insulator in its interior but whose surface contains conducting states [1-6]. Specifically, the prediction and preparation of three dimensions TIs make it of great interesting [7, 8]. Quantum anomalous Hall effect (QAHE) will occur when ferromagnetic order is introduced to this kind of materials [9-11]. In Cr doped Bi_2Se_3, the Curie temperature has an estimated value of 76 K at the doping level of 7.4% [11, 12]. Liu *et al.* observed an enhanced T_c of 50 K in $Bi_{2-x}Cr_xSe_3$ [13]. Ferromagnetism was observed in Cr doped Bi_2Se_3 by Chotorlishvili *et al.* [14], where the magnetic atoms prefer to orientate perpendicular to the TI surface. The antiferromagnetic order will appear by the magnetic impurity dopants of Gd in Bi_2Se_3 [15]. Zhao *et al.* found that Cr atoms adsorption on Bi_2Se_3 can induce a spin-polarization with total net magnetic moments of 2.157 μ_B [16]. Collins-McIntyre *et al.* reported a magnetic hysteresis loop with a coercive field of ~80 Oe and a ferromagnetic transition temperature of 20 K for 5.2% Cr doped Bi_2Se_3 [17]. However, Wang *et al.* showed that the Cr atoms deposition on Bi_2Se_3 surface would not lead to the gap opening at the Dirac point [18]. On the other hand, the electrical field tunning of magnetism was greatly researched in the magnetic topological insulators. Wang *et al.* proposed that electric field could induce the quantum phase transition from ferromagnetism to paramagnetism [19]. Wei *et al.* focused on the $Bi_{2-x}Mn_xSe_3$ crystals topological insulator and found that the concentration of Mn showed metal behavior when $x<0.05$ and it became *p*-type semiconductor at $x=0.07$ [20].

Actually, the observing QAHE still have many problems which have not been solved. For example, the topologically nontrivial band gap opened is too narrow, QAHE can only be found under ~ mK. In

addition, the mechanism of QAHE was still controversial. To figure out these problems, we have studied the magnetic coupling of Cr atoms in Bi_2Se_3 using first-principle calculations and Monte Carlo simulation.

2. Method and Model

2.1. Method

Generally, the magnetic states in magnetic materials can be determined by several competing energies: exchange energy, dipolar interaction, anisotropy energy, Zeeman energy. Therefore, the Hamiltonian of Cr-doped topological insulators can be described by

$$E = -\sum_{ij} J_n \vec{S_i}\vec{S_j} + D[\frac{\vec{S_i}\cdot\vec{S_j}}{r_{ik}^5} - 3\frac{(\vec{S_i}\cdot\vec{r_{ik}})(\vec{S_j}\cdot\vec{r_{ik}})}{r_{ik}^5}] - K\sum_i (\vec{S_i}\cdot\vec{u_i})^2 - H\sum_i S_i^z \qquad (1)$$

Where J_n is the exchange coupling constant, J_1 is the inner layer exchange constant and J_2 is the interlayer layer exchange constant, while J_3-J_7 is the different QLs exchange constant; D is dipolar interaction constant, here $D=0$; K is the anisotropy constant [21, 22]; H is the magnetic field intensity. The magnetization per atom is calculated by $M = <[(\sum_l S_l^x)^2 + (\sum_l S_l^y)^2 + (\sum_l S_l^z)^2]^{1/2}> / N$. We use Monte Carlo (*MC*) to analyze the magnetization reversal processes for Cr doping, where the spins can rotate freely in three dimensions.

2.2. Model

As shown in Figure 1, it is the crystal structure for 3×3×1 Bi_2Se_3 supercell in our VASP calculation. Two Cr atoms are replaced for different Bi atoms and the doping ratio is 3.703%. Projected augment wave (PAW) potentials with Perdew-Burke-Ernzerh (PBE) type of gradient approximation (GGA) is used in the first-principles calculations. Spin-orbital coupling (SOC) is considered. The lattice constants a=4.138 Å and c=28.64 Å for Bi_2Se_3. The cutoff energy was set at 300 eV. The Brillouin zone for the 3×3×1 Bi_2Se_3 is sampled by a gamma-centered 7×7×2 mesh.

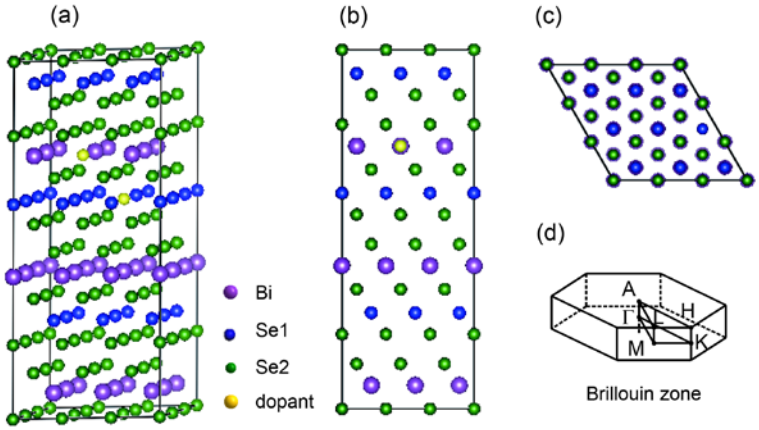

Figure 1. (a) Crystal structure illustration of a 3×3×1 supercell for the modelling of two dopants in bulk Bi_2Se_3 with (b) the side view and (c) the top view. (d) Brillouin zone and high symmetry points of the 3×3×1 supercell.

3. Results and Discussions

3.1. Electronic property of Cr doped Bi_2Se_3

We calculated band structure for Cr doped Bi_2Se_3 with the concentration of 4%, as show in Figure 2. We find an inverted band at the G point due to the strong SOC effect in this material, the opened gap for Cr doped Bi_2Se_3 is 0.08 eV and the magnetic moment is about 3 μ_B. The result shows the magnetic

atoms of Cr can be effectively doped in Bi_2Se_3, ferromagnetic order will appear and the material is still insulator, rendering it to be an ideal material to realize QAHE.

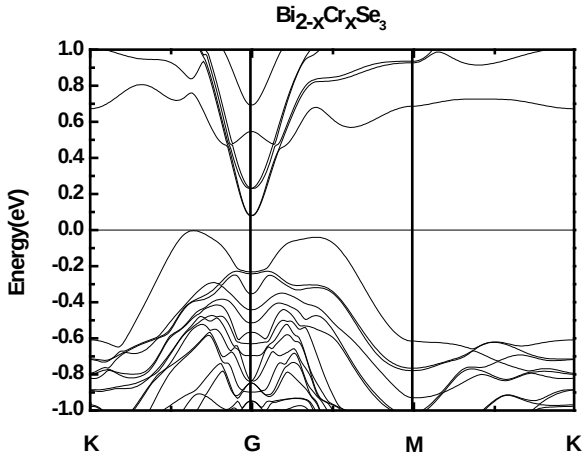

Figure 2. SOC band structure for relaxed Cr doped Bi_2Se_3 concentration of 4% ($x = 0.083$).

3.2. The magnetic coupling of the atoms inner layer

To investigate the magnetic coupling of doped magnetic atoms, two Cr atoms are substituted for Bi atoms, as seen in Figure 1. Our first-principle calculation shows the nearest magnetic coupling strengths is 22 meV, while the strengths after J_7 are less than 2 meV [12]. Therefore, we mainly focus on the distance effect on the magnetic coupling in $Bi_{2-x}Cr_xSe_3$.

As shown in Figure 3, we can find that when the inner layer exchange constant J_1 changes from -1 to 0, the saturation magnetization gradually increased. As inner layer exchange constant J_1 close to zero, the magnetic hysteresis loop is easier saturated. In contrast, when J_1 varies from 0 to 1, the magnetization gradually decreased. What's more, the hysteresis loop arise obviously hysteresis phenomena, and the residual magnetism (M_r) and coercivity (H_c) are not zero. This is due to that system will show antiferromagnetic property when J_1 is negative, accompanying with M_r and H_c close to 0. On the contrary, when J_1 is positive, the material exhibits ferromagnetism with the obviously hysteresis effect emerging. The saturation magnetization (M_s) comes up with a maximum when J_1=0.1.

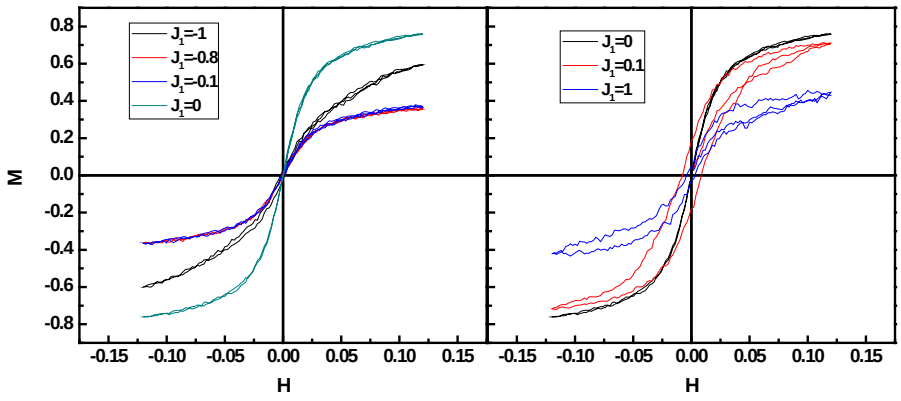

Figure 3. Hysteresis loops of Cr doping Bi_2Se_3 with different J_1 .

3.3. The magnetic coupling of the atoms interlayer layer

From Figure 4, we can see the effect of J_2 is same to J_1. While, M_r and H_c will tend to be zero as J_2 changes from 0 up to 0.2. In addition, M_s, M_r and H_c almost come up to the maximum value at the same time when J_2=0.1, as can be seen in Figure 5.

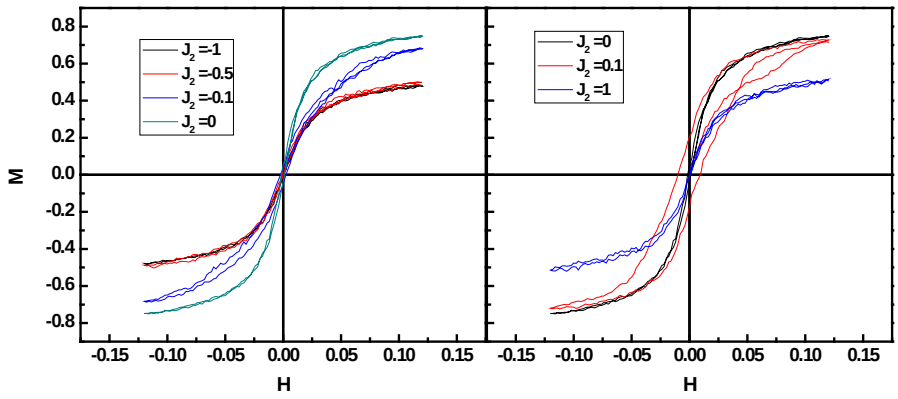

Figure 4. Hysteresis loops of Cr doping Bi_2Se_3 with different J_2.

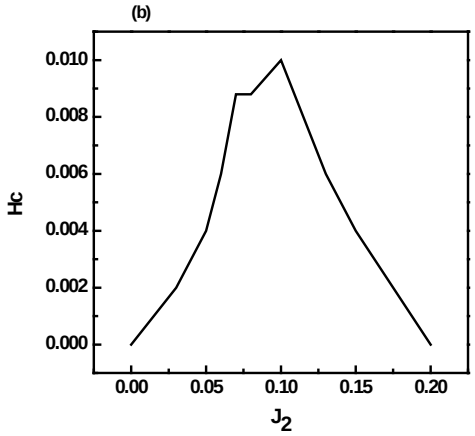

Figure 5. The variation of H_c with different J_2.

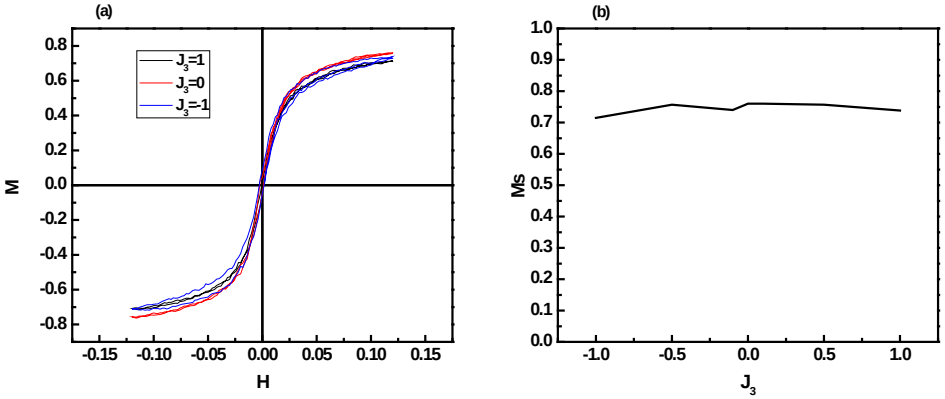

Figure 6. (a) Hysteresis loops of Cr doping Bi_2Se_3 with different J_3, (b) the variation of saturation magnetization M_s with different J_3.

3.4. The magnetic coupling of the atoms in different QLs

Figure 6(a) shows hysteresis loops hardly change when two Cr atoms are doped in different QLs. Therefore, the magnetic coupling between different QLs has little contribution to the system. It is also confirmed in Figure 6(b), the values of M_s and M_r are always at about 0.71 and 0.08, respectively.

Coercivity H_c is always zero. The reason is that the exchange energy is too little as the doping atoms far apart. This result proves that J_3-J_7 have little affection to the system, which is also confirmed by our first-principle calculations [12].

4. Conclusion

In summary, we have studied the distance effect on the magnetic coupling for Cr doped Bi_2Se_3 using first-principles method and Monte Carlo simulation. We found that the hysteresis loops is sensitive to the direct exchange energy between magnetic atoms. When two Cr atoms exist within the same QL, the system shows to be antiferromagnetism when J_1 and J_2 are negative, while ferromagnetism is appear when J_1 and J_2 are positive. However, as the magnetic atoms distribute exceed 1QL, the exchange energies have little contribution to the magnetism. Our results provide important guidelines towards the further efforts of magnetic doping in TIs, in particular, for the realization of QAHE.

Acknowledgments

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