First-principles study of the magnetism of diluted magnetic semiconductors of II-VI type
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The magnetism of the diluted magnetic semiconductors (DMS) receives presently tremendous attention. This attention was born from the widespread vision of semiconductor spintronic devices that promise to revolutionize modern electronics [1]. The interest to the DMS was strongly motivated by the discovery of the ferromagnetism in Ga$_{0.947}$Mn$_{0.053}$As with the Curie temperature ($T_C$) as high as 110 K [2]. Despite intensive experimental efforts the Curie temperature of (GaMn)As could not be raised above 180 K. The design of the DMS materials with the Curie temperature exceeding the room temperature is still an urgent demand. The search for new materials includes both III-V and II-VI systems.

Very recently a (ZnCr)Te sample with high Cr concentration of 20% has been grown [3]. The Curie temperature of Zn$_{0.8}$Cr$_{0.2}$Te was found to be 300 K. This system is the first II-VI DMS where the high-$T_C$ ferromagnetism is reliably detected.

The challenge for the theory is the development of the universal calculational schemes that provide the description of the magnetism of both III-V and II-VI DMS. In Refs. [4] we proposed the method for parameter-free calculation of the exchange interactions and Curie temperature of DMS. The method is based on the density functional theory and does not contain adjustable parameters. Successful applications of the method to the investigation of the electronic and magnetic properties of III-V DMS are discussed in papers [4].

Here we report the application of the method to II-VI DMS. To calculate the exchange interactions in the system the frozen-magnon approach is employed [4]. The Curie temperature is evaluated in the mean-field approximation $k_B T_C = \frac{2}{3} \sum_{j \neq 0} J_{0j}$ where $J_{0j}$ is the Heisenberg exchange parameter between sites $(0, j)$. The calculated exchange parameters for Zn$_{0.75}$Cr$_{0.25}$Te (Fig. 1) reveal strong ferromagnetic interaction between the Cr atoms separated by vector (110). The detailed analysis of the calculational data shows that the exchange interaction is mediated through the Te atoms at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ and $(\frac{1}{2}, \frac{3}{2}, \frac{1}{2})$ which form the exchange path of the system.

![Fig. 1: Heisenberg parameters of the exchange interaction between the Cr moments.](image1)

![Fig. 2: Curie temperature as a function of the band occupation.](image2)
stitution of a Zn atom by a Cr atom creates exactly one hole. The presence of antisites and other defects can decrease the number of holes per Cr atom. To study the dependence of $T_C$ on the number of holes we performed calculations for different band occupations. Figure 2 shows that the decrease of the number of holes leads to fast decrease of the Curie temperature. For number of holes equal to zero the valence band is full and the conduction band is empty. In this case the exchange interaction becomes strongly antiferromagnetic that is reflected in the negative value of the calculated $T_C$. This result supports the physical picture of the antiferromagnetic superexchange mediated by the filled bands and the ferromagnetic kinetic exchange mediated by the holes.

Cardinally different situation is experimentally obtained in another representative of the II-VI DMS: (ZnMn)Se. On the one hand, this system shows a giant Zeeman splitting that reveals strong exchange interaction between the Mn3d states and the semiconductor p states. This property makes (ZnMn)Se useful for the realization of the spin-injection into semiconductor [5]. On the other hand, for Mn concentrations below 7% no magnetic phase transition is observed down to the temperatures much lower than 1 K. This manifests very weak effective Mn-Mn exchange interactions mediated by the semiconductor p states. Our calculations explain this unusual combination of properties. First, calculation of the spin splitting of the electron states at the top of the valence band for a ferromagnetic configuration of the Cr moments gives a large constant of the p-d exchange interaction $J_{pd}$ (Fig. 3). On the other hand, we show that the contributions of different semiconductor states into the Mn-Mn exchange compensate (Fig. 4) that results in a very small interatomic exchange parameters. A large value of the semiconductor gap in ZnSe plays important role in the formation of weak interatomic exchange despite strong p-d interaction.

The account for Hubbard $U$ within local-density-approximation+U (LDA+U) approach gives better overall agreement with experiment for (ZnMn)Se than LDA.

References