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Carrier-mediated ferromagnetism in N co-doped (Zn, Mn)O-based diluted magnetic semiconductors

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Abstract

Mn-doped ZnO is anti-ferromagnetic spin glass state, however, it becomes half-metallic ferromagnets upon hole doping. In this Letter we report a theoretical study of (Zn, Mn)O system codoped with N, and show that this codoping can change the ground state from anti-ferromagnetic to ferromagnetic. We have carried out the first-principles electronic structure calculations and report total energy to estimate whether the ferromagnetic state was stable or not. Our approach is based on the spin-polarized relativistic Korringa–Kohn–Rostoker (SPR–KKR) density functional theoretical (DFT) method, within the coherent potential approximation (CPA). Self-consistent electronic structure calculations were performed within the local density approximation, using the Vosko–Wilk–Nusair parameterization of the exchange-correlation energy functional. Our results for energy difference between ferromagnetic sate and spin glass state as well as their dependence on concentrations were presented and discussed.

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Keywords: First-principle calculations; Ferromagnetism; DMS; ZnO

1. Introduction

ZnO is an attractive semiconductor for solid state blue to UV optoelectronics, including laser development. Transparency to visible light provides opportunities to develop transparent electronics, UV optoelectronics, and integrated sensors, all from the same material system. ZnO is a direct band gap semiconductor with $E_g = 3.2$ eV. Doping with certain transition metal dopants is predicted to yield ferromagnetism. Moreover, zinc oxide is a well-known piezoelectric and electro-optic material, and can be easily deposited in thin film form [1,2].

A significant challenge to the widespread exploitation of ZnO-related materials in electronic and photonic applications is the difficulty in achieving p-type material.

Several authors reported room temperature magnetic hysteresis loops for ZnO doped with Mn, Co or Ni, although others did not find any evidence of ferromagnetism [3,4].

Dietl et al. [5] predicted that Tc can be pushed above room temperature when some 3d transition metals is doped into GaN or ZnO which are two most extensively studied wide band-gap semiconductors because of their applications in blue lasers and optoelectronic devices [6].

The most extensively studied systems are (Ga, Mn)N and (Zn, Mn)O as these were predicted to be ferromagnetic at or above room temperature [1]. As a host material, ZnO has certain advantages over GaN. It is transparent and up to 35% Mn can be doped into ZnO by the pulsed laser technique [7].

The most promising dopants for p-type material are the group V elements. There have been reports suggesting acceptor doping with group V substitution. Photoinduced paramagnetic resonance studies of N-doped ZnO crystals indicate the presence of an acceptor state due to nitrogen substitution [8].

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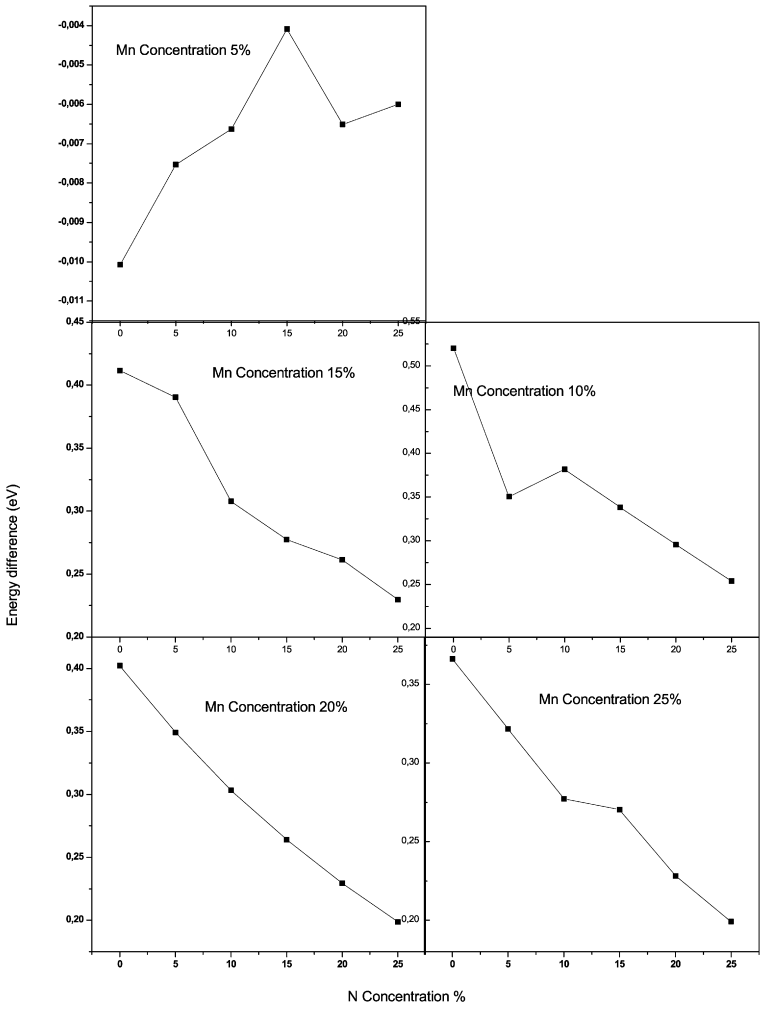

Fig. 1. Total energy differences ΔE (meV) for various Mn doping concentrations x versus N concentration.

In this Letter we report a theoretical study of (Zn, Mn)O system codoped with N, and show that this codoping can change the ground state from anti-ferromagnetic to ferromagnetic.

Sato and Katayama-Yoshida [9], had carried out KKR–CPA calculations in randomly substituted 3d TM impurities in ZnO and found ferromagnetic state to be stable for half-filled or more than half-filled impurities such as V, Cr, Fe, Co, and Ni, while a spin-glass like state is found to be stable for ZnO containing 5% of Mn impurities.

In order to stabilize the FM phase, it is necessary to insert carriers into the system. As ZnO is usually n-type doped with free in the conduction band. The stabilization of ferromagnetism will be more efficient when the carries are holes instead of electrons.

Considerable experimental work is available on the (Zn, Mn)O system. However, the results have been rather controversial. While some groups have reported ferromagnetism in (Zn, Mn)O systems [10,11] others report observations of anti-ferromagnetic or spin-glass behaviour [12]. Similarly conflicting results also exist concerning the distribution of Mn in ZnO. Cheng et al. [13] find Mn to be distributed homogeneously, yet Jin et al. report clustering of Mn atoms [14].

2. Computational details

Within the present work we used the relativistic version of spin-density-functional theory (SDFT) [15,16] based on ab initio method, Korringa–Kohn–Rostoker (KKR)-coherent potential approximation (CPA)-local density approximation (LDA) [17–19]. Self-consistent electronic structure calculations of exchange-correlation potential and energy were performed within the local density approximation, using the Vosko–Wilk–Nusair parameterization of the exchange-correlation energy functional [20].

Mn impurities are introduced randomly into cation sites of the ZnO semiconductor. This substitutional disorder in Diluted Magnetic Semiconductors (DMS) is well described by the Korringa–Kohn–Rostoker coherent-potential approximation (KKR–CPA) method [18]. While the magnetic disorder was included within the disordered-local-moment (DLM) state. Relativistic effects are considered in the scalar relativistic approximation. Throughout the present calculations, we used the SPR–KKR package developed by Ebert [21]. The shape of the crystal potential is approximated by a muffin-tin potential, and the wave functions in the respective muffin-tin spheres were expanded in real harmonics up to $\boldsymbol{l} = 2$, where $\boldsymbol{l}$ is the angular