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Ferromagnetism in alkali-metal-doped AlP: An ab initio study

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ABSTRACT

By using first-principle calculations within the generalized gradient approximation, we investigated the magnetic and electronic properties of Group IA alkali-metal (Li, Na, K)-doped AlP. The main purpose of this study is to determine the effect that different types of alkali dopants have on the magnetic behavior of AlP. The magnetic moment induced by the Li dopant is negligible in $(2 \times 2 \times 2)$ Li-doped AlP supercell. However, it is also found that the magnetism is affected by the Li doping concentration. The Na and K dopants in AlP induce a net magnetic moment of 2.0 μB. Half-metallic characteristics are achieved in K-doped AlP. The calculated density of states and the spin density distribution of Na- and K-doped AlPs show that the magnetism derives from the p–p interaction between the dopants and the neighboring anions. However, the spontaneous polarization cannot occur in Na-doped AlP. The K-doped AlP has the AFM ground state which is the unstable at room temperature. The results reveal that the Na- and K-doped AlPs are unlikely to be potential DMSs for the development of new spintronic devices.

1. Introduction

Diluted magnetic semiconductors (DMSs), which utilize electron spin to create new functionalities beyond those of conventional semiconductors, have been a crucial material for the design of new spintronic devices, such as spin valves, spin light emitting diodes and magnetic sensors [1–4]. For practical device applications, however, robust ferromagnetism with a high Curie temperature is required. Therefore, diluted magnetic semiconductors doped with magnetic transition metal (TM) ions were thought to be the ideal materials for this field. In the past decade, considerable work has been directed towards TM doped III–V semiconductor-based DMSs both theoretically and experimentally, and ferromagnetism above room temperature has been observed [5–8]. However, the origin of this observed ferromagnetism is still under debate. Furthermore, much research has been focused on investigating nonmagnetic ion doping so as to eliminate any problems arising from possible magnetic precipitates in semiconductors. These efforts provide a new research avenue for the development of new classes of high temperature DMSs. Additionally, ferromagnetism in nonmagnetic cation- and anion-doped semiconductors that do not contain magnetic 3d TM ions has been discovered [9–13]. In this respect, alkali metals are theoretically predicted to be potential nonmagnetic dopants for some semiconductors. Ferromagnetism has been reported in alkali-metal doped oxides, nitrides, arsenide and phosphide semiconductors [14–20].

Unfortunately, a report on the magnetic properties of an AlP successfully doped with an alkali metal still does not exist. In this work, first-principle calculations are performed so as to investigate the effect of nonmagnetic alkali dopants on the electronic structure and magnetic properties of alkali-doped AlP. The roles played by Li, Na and K in ferromagnetic properties are investigated in order to establish its DMS characteristics. The paper is organized as follows: the computational detail is outlined in Section 2; the electronic structure and magnetic properties are analyzed in Section 3; and finally, the last section summarizes the results and details the conclusions.

2. Methodology

The calculations were performed using DFT-based full-potential linearized augmented plane wave method [21] implemented by the WIEN2k package. A 64-atom $(2 \times 2 \times 2)$ supercell had been constructed based on the supercell approach [22], and one Al was replaced by an X(Li, Na, K) atom so that the doping concentration is 3.125%. The AlP compound is in space group F-43m. Furthermore, the Al atom is present at (0,0,0), whereas the P atom is at (1/4,1/4,1/4). The radiuses of the muffin-tin spheres (R_{MT}) for the X, Al and P atoms were chosen to be 2.2 a.u., which is large enough to ensure nearly touching spheres as well as minimize the interstitial space. The exchange-correlation potentials were

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constructed using GGA formalism [23]. Additionally, in FP-LAPW calculations, the core states were treated fully relativistically; while for the valence states, a semi-relativistic calculation was performed. The accuracy of the basis set was determined by K_{max}, which is the largest wave vector of the basis set, so the plane wave cut-off parameters were decided by $R_{MT} * K_{max} = 7$. Furthermore, G_{max} for the Fourier expansion of potential in the interstitial region was set as12 a.u. The k-space integration was carried out using the modified tetrahedron method [24], the k-space integration by a mesh of 100 k-points in the full Brillouin zone and with 8 special k-points in the irreducible Brillouin zone (IBZ). Finally, the energy convergence criterion was set as 10^{-5} Ry. The inclusion of all these parameters is sufficient to guarantee properly accurate results.

3. Results and discussion

In order to investigate the origin of the spin polarization (SP), Fig. 1 illustrates the spin-resolved total and the partial density of states (PDOS) of the alkali-doped AlP supercell. A general observation that can be concluded from these figures is that the ground state properties of AlP compound have been significantly modified due to the substitution of the Na and K dopants at the Al site. The Fermi level is shifted across the top of the valence band in comparison to the perfect crystal. The new states were produced at E_F, which accounts for the doping induced magnetism. Furthermore, due to the observed band gap, the doping of group-I elements introduces holes into the valence band; turning the host compound into p-type semiconductor. Fig. 1(a) shows the spin-polarized total density of states (TDOS) of a pure AlP compound for comparison. The valance and conduction band is primarily composed of Al-p and P-p states. For Li-doped AlP, as shown in Fig. 1(b), the top of the valence band is mainly formed by the 3p states of anions, with little contribution from the Al cations. The Li substitution obviously does not contribute to the spin polarization around the top of the valence band. The DOSs of the majority spin and the minority spin are completely symmetric. Therefore, there is no resultant spin polarization in the Li-doped AlP. However, the total DOSs of both the Na- and K-doped AlPs are clearly spin-polarized, as shown in Fig. 1(c) and (d). The minority DOS in vicinity of the Fermi level is significantly modified and split, which endows the 100% spin polarization. The spin-polarized electronic band structures (Fig. 2) show that the Na- and K- doped AlPs depict half-metallic behavior of the minority spin channel. The doped AlPs are excellent half-metallic (HM) ferromagnets with large half-metallic gaps in the spin channels [25]. This suggests that ferromagnetism is possible in a Na- and K-doped AlP. The total and partial DOS of the K-doped AlP are red shifted and away from E_F due to the reduced interaction of the p–p states, as compared to that in the Na-doped AlP. The states at E_F exhibit a mixed character of P-p states and p states of dopants. Al-p states primarily contribute to the bottom of the conduction band (CB) and contribute equally for both the majority and minority channels. The hybridization of the p–p states around E_F results in an HM gap in the minority spin channel. The magnetic moments are mainly formed by the unpaired 3p state of the neighboring anions.

Table 1 shows the total and atomic resolved magnetic moments of the $Al_{31}XN_{32}$ supercell. As indicated in the results, there is almost no magnetic moment in the Li-doped AlP, whereas a Na and K dopant induces a net magnetic moment of 2 μB in the

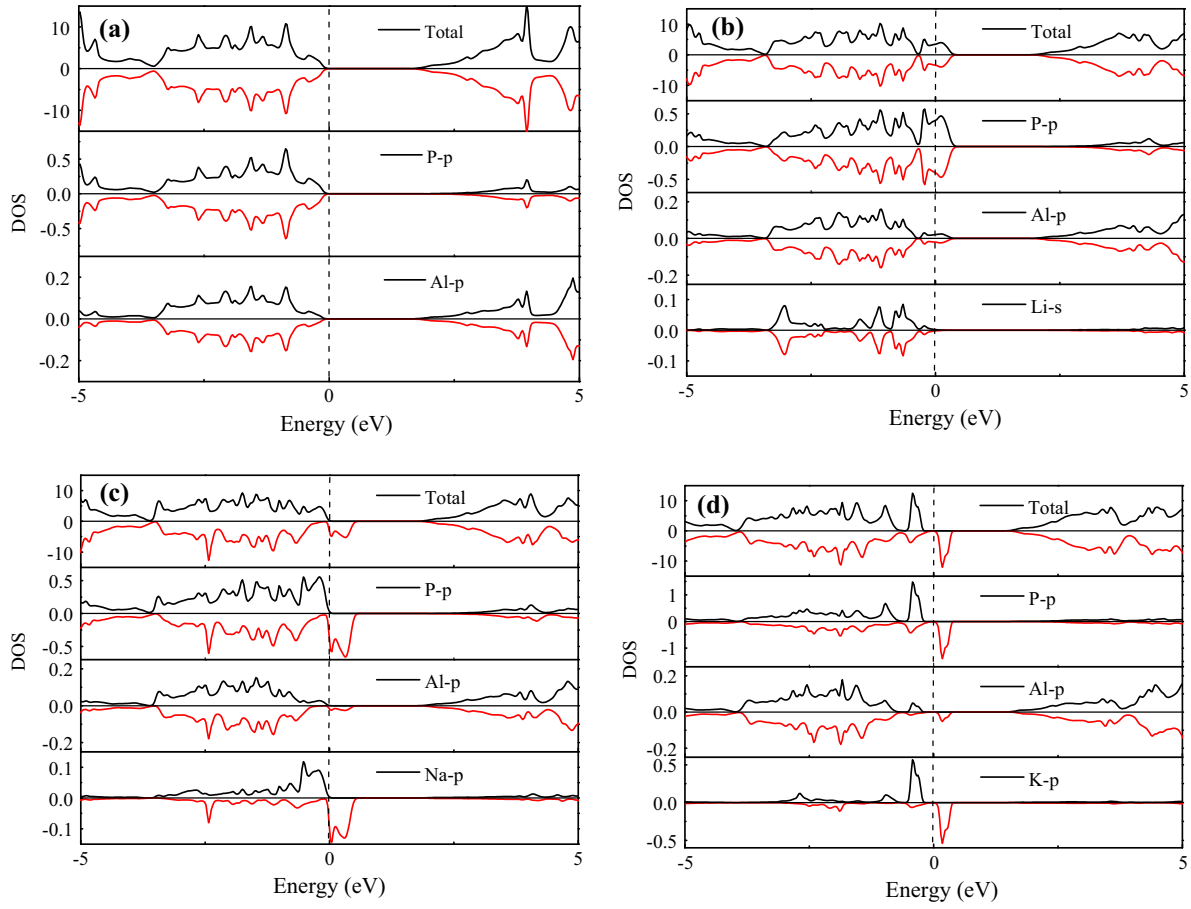

Fig. 1. Totaland partial DOS of (a) $Al_{32}P_{32}$, (b) $Al_{31}LiP_{32}$, (c) $Al_{31}NaP_{32}$, (d) $Al_{31}KP_{32}$. The Fermi energy is indicated by the dashed vertical line. Positive values of the DOS correspond to the majority spin (up) electrons and negative values to the minority spin (down) electrons.