Structural and electronic properties of Eu- and Pd-doped ZnO

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Abstract

Doping ZnO with rare earth and 4d transition elements is a popular technique to manipulate the optical properties of ZnO systems. These systems may also possess intrinsic ferromagnetism due to their magnetic moment borne on 4f and 4d electrons. In this work, the structural, electronic, and magnetic properties of Eu- and Pd-doped ZnO were investigated by the ab initio density functional theory methods based on generalized gradient approximation. The relative stability of incorporation sites of the doped elements in the ZnO host lattice was studied. The ground state properties, equilibrium bond lengths, and band structures of both the ZnO:Eu and ZnO:Pd systems were also investigated. The total and partial densities of electron states were also determined for both systems. It was found that in the ZnO:Eu system, ambient ferromagnetism can be induced by introducing Zn interstitial which leads to a carrier-mediated ferromagnetism while the ZnO:Pd system possesses no ferromagnetism.

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Introduction

Semiconductor metal oxides, with applications in the photoelectrochemical cells, diluted magnetic semiconductors (DMS), field effect transistors, and photoluminescence devices, have recently initiated dynamic research activities [1-3]. In particular, ZnO has a significant advantage for applications in optical [4] and spintronic [5] devices. As a result, doping ZnO with various elements has been a popular technique to manipulate and control ZnO’s extrinsic properties for device applications [6]. Specially rare earth (RE)- and transition metal (TM)-doped ZnO systems exhibit interesting optical and magnetic properties, which do not exist in undoped ZnO. Optically, ZnO systems doped with RE ions have been intensively investigated as electroluminescent systems with wide technological applications [7]. In RE-doped ZnO, the intra-ionic 4f transitions of RE ions form luminescent centers which generate narrow and intense emission lines [8]. While the enhancement in the optical absorption of TM-doped ZnO can transfer these materials to efficient photocatalysts [9].

Magnetically, the intrinsic magnetic moment, borne by the RE and TM ions, makes the RE and TM-doped ZnO systems to be potential diluted magnetic semiconductors with applications in spintronic devices. Over the past decade or so, a considerable amount of effort has been made on searching for ZnO-based DMSs with ferromagnetism at ambient. This goal is meant to be achieved by doping ZnO with mainly the first row TMs, such as Co, Mn, and Fe [10]. However, most recently, interesting magnetism has been observed in other metal oxides doped with the second row TMs, namely in Sn2O3:Pd system [11]. This stimulated further search for possible ferromagnetism [12] and functional optical properties [13] in ZnO:Pd. The realization of magnetism in the ZnO:Pd system is motivated by a previous theoretical prediction [14] and experimental observation [15] of ferromagnetism in Pd clusters. In addition to systems containing TM ions, Eu-doped ZnO (ZnO:Eu) has also shown room temperature ferromagnetic ordering [16] which is partially caused by the high magnetic moment of the Eu ions. In this work, the structural and electronic properties of the ZnO:Pd and ZnO:Eu systems are investigated by a density functional approach. Furthermore, the effects of ZnO’s two dominant point defects [17], Zn interstitial (ZnI) and O vacancy (VO), on the functional properties of these materials are studied. The results of theoretical investigations presented here will shed light on the origin of the functional properties of this relatively new family of materials.
Computational details

*Ab initio* calculations were performed with a density functional theory-based DMOl3 package developed by Accelrys [18,19]. Geometry optimization and partial density of states (PDOS) calculations were performed with “double-numeric plus polarization” (DNP) basis set while generalized gradient approximation (GGA) based on Perdew-Wang formalism was applied for correlation functional [20]. Real-space global cutoff radii were set for all elements at 5 Å. Since only valence electrons would affect the physical properties, the nuclei and core electrons were replaced by DFT semi-core pseudopotentials with a relativistic correction [21]. A Brillouin zone sampling was carried out by choosing the $2 \times 2 \times 2$ k-point set using the Monkhorst-Park scheme with a grid spacing of approximately 0.04 Å$^{-1}$ between k points. The convergence thresholds for energy, Cartesian components of internal forces acting on the ions, and displacement were set to be $10^{-5}$ eV/atom, 0.05 eV/Å, and 0.001 Å, respectively. A convergence testing was performed, first by increasing the k point mesh to $3 \times 3 \times 3$; it was found that the total energy differs less by $10^{-5}$ eV/atom. Then, the k-point mesh was fixed at $2 \times 2 \times 2$, and the cutoff radii were set for all elements to be 5.5 Å. Once again, no significant change in the total energy was obtained. Thus, the results were well converged.

The formation enthalpy and bandgap for undoped ZnO was calculated to be -3.5 and 2.0 eV, respectively. The formation enthalpy is in good agreement with the experimental value of -3.64 eV [22]. However, the bandgap is underestimated by 1.4 eV which is attributed to the GGA intrinsic error. The calculated lattice constants for undoped and fully optimized ZnO were found to be 3.279 Å for $a$ and 5.281 Å for $c$, which are in good agreement with the experimental data [23], overestimated by only 0.9% and 1.5%, respectively. The Zn-O bond lengths in the relaxed structure were 2.005 and 1.997 Å along the $c$ direction and $ab$ plane, respectively. In order to avoid the artificial hydrostatic stress in the doped structures, the lattice parameters were fixed to the calculated values of the undoped ZnO while only the internal atomic coordinates were relaxed.

To simulate the low concentrations of dopants in ZnO, a large supercell of $4a \times 4a \times 2c$ was adopted for calculations. The original supercell contained 64 Zn-O formula units. By introducing one dopant in the substitutional or interstitial site, the concentration of the dopants would be 1.4%. Having the periodic boundary conditions applied, the average separation of the dopant ions is 13.114 Å. This distance is large enough to avoid artificial interaction between the dopants. As a result, the calculations on this supercell will sufficiently resemble the experimental conditions of diluted dopant concentrations. The formation energy ($E^f$) of the dopants or a cluster of dopants in ZnO’s host lattice was calculated as follows:

$$E^f = E^f (\text{ZnO:M}) - E^f (\text{ZnO}) + n\mu_{\text{Zn}} - \mu_{\text{M}} + qE_F \quad (1)$$

in which $E^f$, $\mu$, and $E_F$ represent total energy, chemical potential for respective elements, and Fermi energy, respectively. $\mu_{\text{Zn}}$ and $\mu_{\text{M}}$ are set to be the calculated energies of metallic Zn and the dopants (Pd or Eu) per element. $n$ represents the number of Zn atoms removed from the supercell, which is zero in the case of the interstitial dopant and one for the substitutional dopant. $q$ stands for the net number of electrons transferred from the defect to the conduction band. Since only neutral supercells were adapted for the calculations, $q$ is zero for all configurations.

The ZnO:Eu system

The incorporation mechanism of Eu ions in ZnO’s host lattice was investigated by calculating the $E^f$ of the substitutional Eu (Eu$_{Zn}$) and interstitial Eu (Eu$_{I}$) in the stoichiometric ZnO as presented in Table 1. It was found that the $E^f$ of Eu$_{Zn}$ is -2.391 eV while the $E^f$ of Eu$_{I}$ is 1.429 eV. Such a large difference in $E^f$ indicates that Eu ions favorably substitute Zn ions rather than taking the interstitial sites of the ZnO lattice. The local geometry of the Eu$_{Zn}$ and Eu$_{I}$ is presented in (a) and (b). Figure 1a shows that the length of Eu-O bond along c direction ($ab$ plane) has increased to 2.280 Å (2.260 Å) in the ZnO:Eu$_{Zn}$ system. The increase of the bond lengths is approximately equivalent to an expansion of 14% along the c direction and 13% within the $ab$ plane with respect to the Zn-O bond length in an undoped ZnO. On the other hand, in the ZnO:Eu$_{I}$ system, in which Eu$_{I}$ binds to three Zn and three O ions, the length of the Eu-O and Eu-Zn bonds were found to be 2.323 and 2.857 Å comprising an expansion of 16% and 24%, respectively compared to the unrelaxed structure. In the ZnO:Eu$_{Zn}$ system, although the length of the Eu bonds is also substantially expanded, the expansion is much smaller than that in the ZnO:Eu$_{I}$ system. As a result, the ZnO:Eu$_{Zn}$ system reaches the structural stability with less lattice distortion.

Next, the $E^f$ of Eu$_{Zn}$ in the nonstoichiometric ZnO was studied by considering two distinct situations that lead

| Configuration       | $E^f$ (eV) | Eu’s spin number | Magnetic ground state |
|---------------------|-----------|-----------------|----------------------|
| ZnO:Eu$_{Zn}$      | -2.391    | 6.800           | Paramagnetic         |
| ZnO:Eu$_{I}$       | 1.429     | 6.859           | Paramagnetic         |
| ZnO:Eu$_{Zn}$ + V$_O$ | 1.772     | 6.907           | Paramagnetic         |
| ZnO:Eu$_{Zn}$ + Z$_n$ | 2.776     | 6.879           | Ferromagnetic        |
to Zn excess: first by including a VO in the ZnO:EuZn + VO system and then by including a ZnI in the ZnO:EuZn + ZnI complexes in the ZnO lattice was found to be 1.772 and 2.776 eV, respectively. The local geometry of the Eu ion in the ZnO:EuZn + VO and ZnO:EuZn + ZnI are presented in Figure 1c, d respectively. In the ZnO:EuZn:VO system, the length of the Eu-O bond is 2.280 Å along the c direction and 2.260 Å within the ab plane, which is identical to the bond length in the ZnO:EuZn system without VO. However, in the ZnO:EuZn+ZnI system, the length of the Eu-O bond expanded to 3.517 Å along the c direction and 3.344 Å within the ab plane. Such an expansion corresponds to a 75% and 67% increase in the length of the Eu-O bonds along the c direction and within the ab plane, respectively compared to the unrelaxed structure. As a result, the EuZn + ZnI complex has the highest E_f and lattice distortion.

To investigate the electronic properties of the ZnO:Eu systems, the total and Eu’s 4f partial density of states (DOS) of all configurations were calculated and presented in Figure 2. A general feature of the Eu’s 4f states in all configurations is that Eu’s 4f states are localized in a narrow impurity band of the width of approximately 1 eV, which is located just below the Fermi level. Such localization of the 4f states indicates that 4f electrons are not affected by the local crystal environment. This point is further reinforced by the Eu’s magnetization as presented in Table 1. The spin number (S) of the Eu ions in all configurations is approximately 6.9, very close the spin number of free Eu atoms which indicates the infinitesimal hybridization of Eu’s 5f orbitals with other orbitals in the host crystals. According to Figure 2a, b, in stochiometric systems, ZnO:EuZn and ZnO:EuI, there are minor electronic states available at the Fermi level resulting in limited mobile carriers in those systems. However, this amount of carriers is not sufficient to establish carrier-mediated magnetism in the stochiometric ZnO:Eu systems, and these systems remain paramagnetic [24]. By introducing VO, in the ZnO:EuZn + VO system, the VO’s impurity states appear below Eu’s 4f states as shown in Figure 2c. Thus, VO does not enhance the carrier concentration in the ZnO:EuZn + VO system either. As shown in Figure 2d in the ZnO: EuZn + ZnI system, ZnI’s 4s states appear in a small peak at the Fermi level, partially hybridizing with Eu’s 4f states and introducing further carriers at the Fermi level. To investigate the possibility of ferromagnetic coupling in the defective systems, two substitutional Eu ions were located in the supercells. Then the E_f of each system was calculated once for ferromagnetic magnetic alignment (E_{FM}) and once again for antiferromagnetic magnetic alignment (E_{AFM}) of the Eu ions. Finally, \( \Delta E \) is defined to be \( E_{AFM} - E_{FM} \) which is an indicator of

Figure 1 The relaxed crystallographic structure of the ZnO:Eu systems. Representing (a) ZnO:EuZn, (b) ZnO:Eu, (c) ZnO:EuZn + VO, (d) ZnO: EuZn + ZnI systems. Red, blue, and gray balls represent the O, Zn, and Eu ions, respectively. For each system, the calculated length of the Eu bonds is presented along both the c direction (or [001]) and within the ab (basal) plane.
ferromagnetic phase stabilization. For Eu ions separated by approximately 3.4 Å (nearest possible distance), \( \Delta E \) was found to be 21 meV for the ZnO:Eu + ZnI system and 3 meV for the ZnO:Eu + VO system. However, for both systems, the \( \Delta E \) vanishes when the separation between the Eu ions increases to approximately 6 Å. This trend in \( \Delta E \) indicates that ZnI induces short range ferromagnetic coupling in the ZnO:Eu system.

The ZnO:Pd system

In the ideal ZnO lattice, the length of the O-Zn bond within the basal plane is 1.997 Å. The radius of O\(^{2-}\) being 1.40 Å, leaves enough room for Pd\(^{2+}\) with an atomic radius of 0.64 Å to substitute the Zn\(^{2+}\) ion without significant lattice distortion to create a Pd\(_{Zn}\). Alternatively, Pd\(^{2+}\) can fit in the octahedral interstitial site which is located in the interstitial channel along \( c \) axis. In addition to the octahedral interstitial site, there is a tetrahedral interstitial site in ZnO which has a Zn\(^{2+}\) ion and an O\(^{2-}\) ion as nearest-neighbor atoms, at a distance of about 1.67 Å, (0.833 times the Zn-O bond length along the \( c \) axis). Thus, a Pd ion cannot be placed at this site without severe geometric constraints. In order to determine the preferred site of the Pd ion in the ZnO lattice, the \( E^f \) of Pd for both the ZnO:Pd\(_i\) and ZnO:Pd\(_{Zn}\) systems were calculated. It was found that the \( E^f \) of Pd\(_{Zn}\) and Pd\(_i\) were 0.776 and 1.612 eV. Such a difference results in high concentration of Pd\(_{Zn}\) over the Pd\(_i\) in thermal equilibrium condition in the ZnO:Pd system. Such a finding is in agreement with the reported experimental data that Pd\(^{2+}\) tends to substitute Zn\(^{2+}\) in ZnO [13]. Figure 3a, b show the local geometry of Pd\(_{Zn}\) and Pd\(_i\) in ZnO. For the ZnO:Pd\(_{Zn}\) system, the Pd-O is 2.127 and 2.199 Å along \( c \) direction and within the \( ab \) plane, respectively with approximate expansions of 6% and 10%, respectively compared to the unrelaxed structure. For the ZnO:Pd\(_i\) system, the Pd-O and Pd-Zn bonds have increased to 2.236 and 2.451 Å, respectively with the expansions of 11% and 6% with respect to the unrelaxed structure. Similar to the ZnO:Eu system, in the ZnO:Pd system, Pd\(_{Zn}\) has a lower \( E^f \) and causes less lattice distortion. Electronically, a Mullikan population analysis indicated that both Pd\(_{Zn}\) and Pd\(_i\) are isovalent to the Zn ions, transferring two electrons to neighboring O ions. This implies that Pd’s 4d shell remains fully occupied, thus the Pd ions are not magnetized in the ZnO host lattice which is reflected in zero magnetization of Pd ions in all configurations as presented in
Table 2. As a result, the stochiometric ZnO:Pd system is nonmagnetic.

Similar to the previous section, the effect of VO and ZnI on the ZnO:Pd systems was investigated by calculating the $E_f$ of the PdZn + VO and PdZn + ZnI complexes in ZnO, which was found to be 6.289 and 3.831 eV, respectively. In the ZnO:PdZn + VO system, the Pd-O bond is 2.300 and 2.358 Å along the $c$ direction and the $ab$ plane, respectively, having 15% (18%) expansion in bond lengths with respect to the unrelaxed structure. In the ZnO:PdZn + ZnI system, the lengths of the Pd-O bond along the $c$ direction and within the $ab$ plane are 3.332 and 3.398 Å, respectively which comprise an expansion of 66% along the $c$ direction and an expansion of 70% within the $ab$ plane compared to the unrelaxed structure.

To investigate the electronic properties of the ZnO:Pd systems, the total and partial density of states of both systems were calculated and presented in Figure 4. As in Figure 4a, the 4$d$ states of PdZn and O’s 2$p$ states hybridize and form bonding ($t^b$) states in the valence band. The antibonding 4$d$ states with $e$ symmetry are located above the $t^b$ states, separated by a gap of approximately 1 eV from the valance band maximum and positioned just below the Fermi Level. The position of the PdZn antibonding states with respect to the Fermi level indicates an n-type behavior of the ZnO:PdZn. Notably, the 4$d$ states are split into bonding and antibonding states. The bonding states with the $t^b$ symmetry hybridize with O’s 2$p$ orbitals along the valance band. The antibonding states with $e$ symmetry are divided into three peaks in the fundamental bandgap region. This is the major difference of the DOS of the Pd$_d$ and Pd$_Zn$, which may be caused by the different crystal fields experienced by each site. The closely located peaks in the bandgap region results in the high optical activity of the ZnO:Pd$_d$ system, in particular, they may be the mechanism behind the observed red shift in the photoluminescence.

Table 2. The $E_f$, Pd’s spin number, and the magnetic ground state of the ZnO:Pd$_{Zn}$, ZnO:Pd$_d$, ZnO:Pd$_{Zn} + VO$, and ZnO:Pd$_{Zn} + ZnI$ are presented.

| System         | $E_f$ (eV) | Pd’s spin ($\frac{\hbar}{2}$) | Magnetic ground state |
|----------------|-----------|-------------------------------|-----------------------|
| ZnO:Pd$_{Zn}$ | 0.776     | 0.000                         | Nonmagnetic           |
| ZnO:Pd$_d$    | 1.612     | 0.000                         | Nonmagnetic           |
| ZnO:Pd$_{Zn} + VO$ | 6.289 | 0.000                         | Nonmagnetic           |
| ZnO:Pd$_{Zn} + ZnI$ | 3.831 | 0.000                         | Nonmagnetic           |
spectrum in the ZnO:Pd system [13]. The position of the $e$ states from the Fermi level and the absence of exchange splitting indicate that PdI is an $n$-type dopant with no magnetization. In nonstochiometric systems, ZnO:Pd$^{+}\text{Zn}$ + V$_{O}$ and ZnO:Pd$^{+}\text{Zn}$ + Zn$_{I}$, the Pd's 4$d$ states, as shown in Figure 4c, d, are distributed in a similar pattern to the ZnO:Pd$^{+}\text{Zn}$. It exhibits the Pd's spin-up and spin-down states which are fully degenerate, indicating the absence of any magnetization per Pd ion. Therefore, these systems are nonmagnetic even when V$_{O}$ and Zn$_{I}$ exist in the ZnO:Pd system.

**Conclusion**
The structural and electronic properties of the ZnO:Eu and ZnO:Pd systems were investigated by *ab initio* techniques. It was found that both Eu and Pd ions substitute Zn sites in the ZnO host lattice favorably. With Zn excess in the ZnO:Eu system, the carrier-mediated ferromagnetism can be induced by Zn$_{I}$. In the ZnO:Pd system, the Pd ions prefer to substitute Zn sites, forming substitutional Pd. Additionally, the Pd ions are isovalent to Zn ions and, consequently, their 4$d$ shell remains fully occupied with no magnetization per Pd ion.

**Abbreviations**
DFT: density functional theory; DMS: diluted magnetic semiconductors; DNP: double numerical polarized; DOS: density of states; GGA: generalized gradient approximation; PDOS: partial density of states.

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**Authors' contributions**
MHNA performed the computational work and drafted the manuscript. YBZ, RKZ, SPR, and SL jointly developed the model and co-drafted the manuscript. All authors read and approved the final manuscript.

**Competing interests**
The authors declare that they have no competing interests.

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