Anisotropic ferromagnetism in carbon doped zinc oxide from first-principles studies

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A density functional theory study of substitutional carbon impurities in ZnO has been performed, using both the generalized gradient approximation (GGA) and a hybrid functional (HSE06) as exchange-correlation functional. It is found that the non-spinpolarized C_{2\text{a}} impurity is under almost all conditions thermodynamically more stable than the C_{\text{O}} impurity which has a magnetic moment of 2\mu_B, with the exception of very O-poor and C-rich conditions. This explains the experimental difficulties in sample preparation in order to realize d^{5}-ferromagnetism in C-doped ZnO. From GGA calculations with large 96-atom supercells, we conclude that two C_{\text{O}}-C_{\text{O}} impurities in ZnO interact ferromagnetically, but the interaction is found to be short-ranged and anisotropic, much stronger within the hexagonal ab-plane of wurtzite ZnO than along the c-axis. This layered ferromagnetism is attributed to the anisotropy of the dispersion of carbon impurity bands near the Fermi level for C_{\text{O}} impurities in ZnO. From the calculated results, we derive that a C_{\text{O}} concentration between 2\% and 6\% should be optimal to achieve d^{5}-ferromagnetism in C-doped ZnO.

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I. INTRODUCTION

Ferromagnetism is a quantum phenomenon which arises from the long range ordering of interacting magnetic moments in a solid. The richness of magnetic properties in solids is due to hybridization of d- and f-orbitals which is decisive for magnetic moment formation and the way individual moments interact with each other in the solid. This is one of the reasons why magnetic semiconductors or diluted magnetic semiconductors (DMS) are traditionally conceived as semiconducting materials with transition metal ions as dopant. Numerous studies of DMS, both experimental and theoretical (see, e.g. Refs. [1,2]) have come up with varying conclusions. In ZnO, substituting the cation by an isovalent transition metal, e.g. cobalt, introduces magnetic moments, but in itself does not lead to ferromagnetic coupling between these moments [3]. Hence, complex strategies, for instance co-doping, and elaborate growth techniques seem to be necessary to fabricate a DMS based on ZnO as host material.

Recently there are reports on magnetism in semiconducting materials without any d- or f-elements [6,9]. This is commonly known as d^{0}-magnetism and discussed for different classes of materials, starting from pure carbon based materials like graphite, graphene, and non-metallic nanoparticles [6,10,11]. The common feature in this class of materials is that magnetism involves the 2p orbitals. The origin of magnetic moment is attributed to incomplete cancellation of majority and minority spin electronic contribution due to the Hund’s type exchange. Magnetic moments have been reported for almost all ionic semiconductors in the presence of intrinsic defects, such as cation vacancies, or in the presence of impurities related to anion substitution. In the following we discuss reports of magnetism in semiconductors with impurities at the anion site.

The formation of a localized magnetic moment, which is a pre-requisite for d^{5}-magnetism, depends on the relative strength of electronegativity between the dopant element and the anion of the host semiconductor. If the bond between the dopant and the cation is weaker than the native bond of the semiconductor, this leads to localized atomic-like 2p orbitals of the dopant and a stable spin-polarized state. On the other hand, if the strength of the bond is stronger than the native bond of the semiconductor, this will cause delocalization of the dopant 2p orbitals due to strong hybridization with the cations. Consequently, there is reduced or vanishing spin-polarization in the system [12]. Hence, substituting the anion site by an element with smaller electronegativity can introduce spin-polarized states in the gap which can then medi-
ocate the magnetic interaction through the double-exchange mechanism as discussed by Mavropoulos et al. 13.

By means of DFT calculations, Peng et al. 14 have shown that many elements in the second row of the periodic table (X = B, N, C) when substituted at the anion site can develop a spin-polarized solution in AlN (XN) and ZnO (XO). However, the nature of the magnetic correlation in case of BO and NO is still under debate 14 17. Specifically for ZnO, Adeagbo et al. found that both NO and NZn develop spin-polarization with almost 1 B/N in ZnO 18. Lyons et al. have studied NO in ZnO by hybrid-functional calculations and observed that NO defects create a deep impurity state 19. These states, if partially filled, can be spin-polarized. Also for CO, DFT calculations predicted a magnetic moment for the neutral or singly charged impurity 20. There are experimental reports of room temperature ferromagnetism in carbon-doped ZnO films grown by pulsed-laser deposition 21 22. Measurements of the anomalous Hall effect have shown that the ferromagnetism is mediated by the charge carriers in ZnO. Remarkably, results have been reported both for samples showing n-type conductivity 21 or p-type conductivity 23. Moreover, Yi et al. reported on room-temperature ferromagnetism in carbon-doped ZnO films when co-doped with nitrogen 24.

In this work, we select ZnO as a host material for the study of d⁰ magnetism. Impurities and defects in ZnO have been an active topic of research 25 26. Here we focus on the magnetic properties of ZnO with carbon as substitutional impurity (on the zinc site denoted by CZn) and on the oxygen site denoted by CO) studied with the help of density functional theory (DFT) calculations. The paper is arranged as follows: In the next Section we present technical details of our DFT calculations. The Results and Discussion Section discusses the stability of carbon impurities in ZnO by calculating their formation energies. The distance-dependent exchange interactions of CO in ZnO are obtained from the total energy differences of ferromagnetic and antiferromagnetic states. Finally, we conclude on the possible connection between C impurities and the ferromagnetism in ZnO found in experiments.

II. COMPUTATIONAL DETAILS

Electronic structure calculations are performed using the plane-wave pseudopotential DFT method as implemented in the Vienna ab-initio simulation package (VASP) 27 28. For studying a substitutional impurity in ZnO, the system is modeled as a supercell consisting of periodic repetitions of the primitive wurtzite cell. Since we use different sizes of supercells, we stick to a particular nomenclature in our discussions. We refer to the size of the supercell with periodic extension nx×ny×nz as “Snxny nz”. The electronic structure of homogeneously distributed impurities has been studied with the relatively small supercell S222, see Fig. 1(c), while to study impurity pairs in ZnO we employ two kinds of large supercells, namely S622 and S226 (Fig. 1(d) and (e), respectively. This is necessary as the wurtzite crystal structure is non-centrosymmetric and the bond lengths are different along the hexagonal plane and along the c-direction. Essentially, S622 intends to scan the interactions along the hexagonal plane of the lattice and the S226 scans the interaction along the c-axis of the lattice.

The calculations have been performed using a plane-wave energy cutoff of 400 eV. We have used the generalized gradient approximation (GGA) with the PW91 parameterization 29 for the exchange-correlation potential and the ion-electron interactions are treated with the projector-augmented wave (PAW) method 30.
The lattice constants of the primitive lattice of ZnO is taken to be $a = 3.29$ Å, $c/a = 1.606$ and the relative shift of Zn and O planes, $u = 0.380$, which were found to be the minimum energy lattice parameters in the GGA calculations [32]. The doped supercell configuration is relaxed to avoid any strain until the structural energy is converged to $10^{-6}$ eV. At this level of tolerance, the forces in the system are found to be below $3 \times 10^{-4}$ eV Å$^{-1}$. The relaxation is performed with a Γ-centered Monkhorst-Pack $k$-points grid of $3 \times 5 \times 5$ and $5 \times 5 \times 3$ for supercells S622 and S226, respectively.

Part of these results have been cross-checked with the full-potential local orbital (FPLO) minimum-basis code, which uses localized basis sets [31]. DFT within GGA gives reliable structural parameters for ZnO but has limitation in predicting the right optical band gap, which is with a value of 0.69 eV grossly underestimated in comparison to the experimental value of 3.4 eV [32]. In addition, the localization of Zn $d$-bands is not properly described by GGA because the hybridization of Zn $d$ and O $p$ bands is overestimated. Thus, the Zn $d$ bands are located almost 3 eV too close to the Fermi level as compared to experimental observation [33]. One way to overcome the problem is through the introduction of hybrid functionals where the electronic exchange potential is mixed with some percentage of Hartree-Fock exchange, while the electronic correlation potential is entirely taken from the GGA. We have employed the screened hybrid-functionals to ascertain the electronic structure of a single impurity. The hybrid-functionals are of the type suggested by Heyd, Scuseria and Ernzerhof [34], with the the exact (Hartree-Fock) exchange and the GGA exchange in the ratio 1:3 [35]. We have used the HSE06 functional for our studies [36].

### III. RESULTS AND DISCUSSION

The stability of an impurity configuration can be judged from its formation energy. Since substitution changes the number of atomic species, the formation energy must be calculated with reference to thermodynamic reservoirs characterized by their chemical potentials. In practice, the values of these chemical potentials reflect the experimental conditions during sample preparation. The formation energy, $E_f$, is thus defined as

$$E_f = E_{\text{Tot}} - E_{\text{ZnO}} - \sum_i n_i \mu_i,$$

where $E_{\text{Tot}}$ is the total energy of the supercell with impurity, $E_{\text{ZnO}}$ is the energy of pure ZnO host with the equivalent number of stoichiometric ZnO units, $\mu_i$ is the chemical potential of a species and $n_i$ is the change in number of the corresponding species in the supercell. In thermodynamic equilibrium with bulk ZnO, the condition

$$\mu_{\text{ZnO}} = \mu_O + \mu_{\text{Zn}}.$$

must always be satisfied. This leaves us with just one unknown, which we choose to be $\mu_O$. However, an interval for the values of $\mu_O$ corresponding to physically meaningful growth conditions can be provided: The upper bound of the chemical potential, corresponding to O-rich growth conditions, is taken from the oxygen molecule, $\mu_O = \frac{1}{2}E_{\text{O}_2}$. Oxygen-poor conditions (or, equivalently, Zn-rich conditions) correspond to the equilibrium with metallic bulk Zn, $\mu_{\text{Zn}} = E_{\text{bulk Zn}}$, and hence, from Eq. (2), $\mu_O = \mu_{\text{ZnO}} - E_{\text{bulk Zn}}$.

#### A. C$_{\text{Zn}}$ and C$_{\text{O}}$ impurities

The formation energies of carbon at the Zn-site (C$_{\text{Zn}}$) and at the O-site (C$_{\text{O}}$) obtained from calculations with a $3 \times 3 \times 3$ supercell of ZnO are summarized in Table 1 for two different environments. The limiting chemical potentials of the C-rich and for C-poor cases are taken as $\mu_C = E_C$ and $\mu_C = E_{\text{CO}} - \frac{1}{2}E_{\text{O}_2}$, respectively, as described in Ref. [20]. We note that the deviation of the values as compared to Ref. [20] is due to the different exchange-correlation functionals used in both calculations. The values obtained by Tan et al. [37] should not be directly compared to ours because they have used the total energies of single atoms as reference chemical potentials. In Fig. 2 the formation energies which we obtain for C$_{\text{O}}$ and C$_{\text{Zn}}$ impurities are plotted as a function of oxygen chemical potential. Our formation energies suggest that C$_{\text{O}}$ and C$_{\text{Zn}}$ impurities are energetically favorable only under carbon-rich conditions (negative values in the formation energy). Also, the formation energy of C$_{\text{Zn}}$ is smaller than the formation energy of C$_{\text{O}}$ in all chemical environments.

We find that the formation of C$_{\text{O}}$ is energetically favorable only in a narrow range of $\mu_O$ shown as the thick (blue) vertical line. This tiny region corresponds to an equilibrium with metallic zinc, i.e., extremely O-poor conditions. Hence stabilizing the C$_{\text{O}}$ defect in ZnO is difficult under common experimental growth conditions of high oxygen pressure where $\mu_O$ approaches the value corresponding to molecular oxygen.

A single C$_{\text{O}}$ in a ZnO supercell leads to a spin-polarized solution with an integral magnetic moment of $2\mu_B$/C as discussed in the literature previously [12, 21, 38]. The spin polarized solution is lower in energy than the non-spin polarized one by
and C\textsubscript{O}\textsuperscript{+} method is shown in Fig. 3 (a), (b) and (c). In contrast, the DOS of pure ZnO obtained from each line (blue colored line) shows the narrow range where the C\textsubscript{O} impurity can compete with C\textsubscript{Zn}.

![Graph showing formation energy of C\textsubscript{O} and C\textsubscript{Zn} as a function of \(\mu\textsubscript{O}\). The thick vertical line shows the narrow range where the C\textsubscript{O} impurity can compete with C\textsubscript{Zn}.](image)

Figure 2: (Color online) The formation energy of C\textsubscript{O} and C\textsubscript{Zn} defects as a function of \(\mu\textsubscript{O}\). The thick vertical line (blue colored line) shows the narrow range where the C\textsubscript{O} impurity can compete with C\textsubscript{Zn}.

A difference of 0.215 eV. The corresponding DOS of C\textsubscript{O} as single impurity in supercell S222 is shown in Fig. 3 (d), (e) and (f), employing the GGA, GGA+\(U\textsubscript{d}\) and HSE06 functionals. For comparison, the DOS of pure ZnO obtained from each method is shown in Fig. 3 (a), (b) and (c). In GGA+\(U\textsubscript{d}\), the value \(U\textsubscript{d} = 7.5\) eV is applied to the (fully occupied) Zn d-orbitals. We find a magnetic solution for C\textsubscript{O} in all the methods. Note that both in GGA+\(U\textsubscript{d}\) and in HSE06, the impurity band in the minority spin is clearly separated from the valence band. The real-space plot of the effective spin density (from GGA) obtained by subtracting the charge densities of majority and minority spins (\(\Delta \rho = \rho\uparrow - \rho\downarrow\) is shown in Fig. 3. One finds that the effective spin density is centered on the C atom and extends to the nearest neighbor Zn atoms and the next-nearest neighbor O atoms with a total magnetic moment of 2\(\mu\textsubscript{B}/C\). We note that the HSE06 functional gives a large exchange splitting of the impurity states and larger band gap for ZnO than GGA, and the impurity states introduced by C\textsubscript{O} which mainly consist of carbon states are deep in the gap when measured from the valence band maximum (VBM). Therefore, it is not possible to use C\textsubscript{O} as an acceptor that could introduce hole states for thermally activated p-type conductivity in ZnO. On the other hand, the more stable C\textsubscript{Zn} impurity acts as a donor and does not lead to any spin polarization, in agreement with Ref. [9].

Finally, we discuss possible compensation effects that could alter the magnetic properties of C\textsubscript{O}. Using DFT studies, Li et al. have shown that the presence of oxygen vacancy (V\textsubscript{O}) quenches the spin-polarization from C\textsubscript{O} in ZnO. Comparing the formation energy of neutral defects, we conclude that both, V\textsubscript{O} and V\textsubscript{Zn}, have positive formation energy, with the formation energy of V\textsubscript{O} being lower than the formation energy of V\textsubscript{Zn} in O-poor conditions (favorable conditions for C\textsubscript{O} as

| carbon rich | carbon poor |
|-------------|-------------|
| O-poor, C\textsubscript{O} | 4.558 | 4.070 |
| C\textsubscript{Zn} | 4.576 | 4.052 (4.0 Ref. [37]) |
| O-rich, C\textsubscript{O} | -1.569 (0.4 Ref. [20]) | 7.059 (9.4 Ref. [20]) |
| C\textsubscript{Zn} | -7.564 (-6.2 Ref. [20]) | 1.064 (2.8 Ref. [20], 2.2 Ref. [37]) |

Table I: Formation energy of C\textsubscript{O} and C\textsubscript{Zn} calculated from Eq. 1. The data show that the C\textsubscript{Zn} impurity is thermodynamically more stable than the C\textsubscript{O} impurity.
seen above). Hence, the role of defects like V_{Zn} and V_{O} can be safely ruled out in contributing to magnetism in the material. However, the magnetic moment of C_{O} can be quenched by H impurities in ZnO that act as donor. We have tested co-doping of H and C in S622 and find that the electron from H compensates one Bohr magneton of magnetic moment. Upon adding two H in the supercell, the magnetic moment is completely quenched to zero. This observation is independent of the distance of the hydrogen from the carbon site in the supercell. In experiments the hydrogen concentration during ZnO synthesis is difficult to control. It has been found that the energy barrier for the escape of H through the ZnO surface can be as large as 0.58 eV [42]. Thus even if one succeeds with C_{O} doping, one may still fail to obtain magnetic moments due to the ubiquitous presence of hydrogen. Similar conclusions have also been derived from formation energy studies [44].

**B. Fixed spin moment calculations using supercell approach**

Although the thermodynamic stability considerations of the previous section point to the difficulty in incorporating larger amounts of carbon on the oxygen site in ZnO, we continue to discuss the consequences of C_{O} impurities for the magnetic properties, assuming that an appreciable concentration can be built up by non-equilibrium preparation techniques. In order to investigate impurity interactions at various concentrations, we perform DFT calculations with both VASP and FPLO using a single C_{O} impurity in the different supercells shown in Fig. 4(a) to (c). These cells correspond to carbon concentrations of 12.5% (for both S122 and S221) and 6.25% (S222). With decreasing the C concentration from to 12.5% and 6.25%, the interaction between impurities becomes weaker and the impurity band becomes narrower. Even though the supercells S221 and S122 have the same doping concentration of 12.5%, they do not lead to equivalent electronic structure because of the anisotropy of the wurtzite crystal. From Table II we note that the magnetic moment of S122 is a fractional value in units of $\mu_B$, an indication of strong hybridization between the C 2p orbitals and the valence band, leading to metallic behavior as shown in Fig. 4(a). However, the magnetic moment of S221 and S222 is integer (2 $\mu_B$), i.e., the system is a magnetic half metal (see Fig. 4(b) and (c)). We carried out fixed-spin-moment (FSM) calculations to test the stability and characteristics of the spin-polarized solutions itself. The plot in Fig. 5 shows the magnetic polarization energy of the supercells shown in Fig. 4(a) to (c). Here the magnetic polarization energy is defined as the energy of the system at a given fixed magnetic moment (m) relative to the energy of non-magnetic solutions (m = 0). When a spin-polarized solution is favored, then the magnetic polarization energy takes negative values. For metallic systems (supercell S122), the minimum magnetic polarization energies are found at non-integer values of m as shown in Table II and the magnetic polarization energies show a smooth parabolic change around the minima. For supercells S221 and S222 the minimum magnetic polarization energy is obtained at an in-
than the values we have estimated from GGA. Consequently, ZnO may tolerate an even larger C concentration before becoming metallic than the values we have estimated from GGA.

The total magnetic moment ($M_{\text{Total}}$) is shown per supercell, while the magnetic moments of Zn ($M_{\text{Zn}}$) and O ($M_{\text{O}}$) are shown per atom, averaged over the nearest neighbor Zn and O, respectively.

| Supercell | $r_{\text{nn}}$ (Å) | $r_{\text{nnn}}$ (Å) | $M_{\text{Total}}$ ($\mu_B$) | $M_{\text{C}}$ ($\mu_B$) | $M_{\text{Zn}}$ ($\mu_B$) | $M_{\text{O}}$ ($\mu_B$) | $E_f$ (eV) |
|-----------|----------------------|-----------------------|--------------------------|---------------------|---------------------|---------------------|-----------|
| S122      | 3.290                | 6.580                 | 0.953 (1.194)            | 0.300 (0.743)       | 0.049 (0.072)       | 0.0465 (0.061)       | -4.009 |
| S221      | 5.284                | 6.580                 | 2.000 (2.000)            | 0.591 (1.235)       | 0.118 (0.076)       | 0.0085 (0.081)       | -3.758 |
| S222      | 6.580                | 10.57                 | 2.000 (2.000)            | 0.584 (1.235)       | 0.118 (0.045)       | 0.0068 (0.058)       | -3.910 |

Figure 6: (Color online) Magnetic polarization energy ($E_{\text{sp}} = E_{\text{nsp}}$) ($sp = \text{spin polarized and nsp = non-sp polarized}$) as a function of the magnetic moment for different supercells from fixed-spin-moment (FSM) calculations. The minima in the magnetic polarization energy indicate the most stable magnetic moment in the supercell. The magnetic polarization energies obtained from VASP and that of FPLO are in close agreement with each other.

Table II: Nearest neighbor (nn) and next-nearest neighbor (nnn) distance of the periodic image of the C impurity in Å, magnetic moments as obtained from VASP (FPLO), and formation energy ($E_f$) for O-poor and C-rich conditions. The total magnetic moment ($M_{\text{Total}}$) is shown per supercell, while the magnetic moments of Zn ($M_{\text{Zn}}$) and O ($M_{\text{O}}$) are shown per atom, averaged over the nearest neighbor Zn and O, respectively.

Next, we study the magnetic interaction between the carbon impurities as a function of their separation. We employ the relatively large supercells S622 and S226 (Fig. 1d and e) to include long-range interactions. The concentration of carbon in these supercells is 4.17% which is within the range of experimental impurity concentrations [21]. As long as the carbon impurities are well-separated (maximum separation of 16 Å in c-direction and 11 Å in ab-direction for S226 and S622, respectively), each impurity retains its magnetic moment of $2\mu_B$ independent of the distance. Therefore, it is reasonable to map the interaction between two C$_\text{O}$ impurities onto an effective Ising Hamiltonian

$$H(r_{i0}) = -\frac{1}{2} \sum_{\langle i \rangle} J_{i0}(r_{i0}) \sigma_i \sigma_0 .$$

The index $i$ runs over the nearest neighbor sites appropriately taking into account the periodic boundary conditions used in the DFT calculations. Without loss of generality we can choose $\sigma_0 = \pm 1$ and $\sigma_i = 1$ to calculate the individual interaction. For a given geometry $r_{i0} = |r_i - r_0|$, the $J_{i0}$ can now be accessed from the total energy difference between the ferromagnetic and antiferromagnetic alignment of the moments of C$_\text{O}$ impurities.

These $J_{i0}$ are plotted in Fig. 7. As a guide to the eye, we include a shaded area bounded by two exponentially decaying functions within which all the interactions are contained. We note that the $J_{i0}$ are positive in the whole range, i.e. the spin interaction between C$_\text{O}$ is ferromagnetic in the host material. Averagely, the $J_{i0}$ decrease with increasing separation $r_{i0}$ between the impurities while there is a large spread in the data for small distances. We categorize the geometric configuration of the impurities using the crystallographic direction and select those configurations that have the major component of their distance vectors in the hexagonal plane (ab-direction). The selected configurations (A1, A3, A4, A6 and A12) are shown as filled symbols in Fig. 7. We observe a strongly anisotropic behavior of $J_{i0}$: the filled symbols are located in the upper part of the shaded area, whereas the...
action can therefore be explained from the band structure of C\textsubscript{O} in ZnO. In the inset of Fig. 7 the band structure of the minority spin states of C\textsubscript{O} in S333 is shown. Since the spin-majority impurity bands are completely filled and thus cannot give rise to any energy gain, only the minority-spin bands can be responsible for the magnetic interactions. We find that the carbon bands are crossing the Fermi level mostly along the hexagonal \textit{ab}-plane, i.e., along the A–L, \Gamma–M, A–H and \Gamma–K directions, while along the \textit{c}-axis (M–L, \Gamma–A and K–\textit{H}) the Fermi energy lies in the gap between the occupied and the unoccupied bands. Therefore, the ferromagnetic interaction is mediated more effectively within the hexagonal \textit{ab}-plane, giving rise to anisotropic ferromagnetism.

We note that the ferromagnetic interaction is rather short ranged (below 10 Å). Ferromagnetism at finite temperatures can thus only be expected at a carbon concentration larger than 2\% (refer to the scale of the C concentration in the upper axis of Fig. 7). Together with the estimate given in Section III.B which gives the upper bound for C concentration to avoid metallic state, we conclude that a C\textsubscript{O} concentration between 2\% and 6\% should be optimal to obtain ferromagnetism due to homogeneously distributed impurities. Moreover, we note that C impurities have a tendency to cluster which poses an upper limit to the useful concentration. As pointed out in previous work \cite{47}, the C impurities tend to form energetically more stable C\textsubscript{2} molecules. In this case, the \textit{ppp}\textsuperscript{*} orbital of the C\textsubscript{2} molecule resonates with the conduction band of ZnO which may lead to ferromagnetism mediated by the host material in n-type ZnO. Thus, this mechanism differs from the ferromagnetic interaction between C\textsubscript{O} impurities described in the present work which originates from conductivity due to impurity bands, and is hence independent of host carriers.

IV. CONCLUSIONS

We have studied the two possible substitutional carbon impurities in ZnO (C\textsubscript{O} and C\textsubscript{Zn}) and find that C\textsubscript{Zn} is energetically more favorable than C\textsubscript{O}. However, C\textsubscript{O} can be stabilized under specific (O-poor and C-rich) growth conditions. This type of environment is uncommon in usual ZnO growth, but could be generated by oxidizing metallic zinc in the presence of an atomic carbon source. Given that a material with a C\textsubscript{O} concentration of 2–6\% can be realized, this would be an interesting prototypical system for \textit{d}\textsuperscript{0} magnetism: Our DFT calculations indicate that C\textsubscript{O} in this range of concentrations is associated with a localized magnetic moment of 2 \mu\textsubscript{B}/C. Partially filled impurity bands
in the minority spin channel mediate ferromagnetic interaction between the CO impurities. These interactions are short-ranged and anisotropic, being stronger within the ab-plane of the wurtzite ZnO crystal than along the c-axis. Based on our calculations, we predict that such layered ferromagnetism could be used as an experimental hallmark to distinguish carbon-induced magnetism from possible other forms of defect-induced ferromagnetism in ZnO.

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