Energetics and magnetism of Co-doped GaN(0001) surfaces: A first-principles study

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Abstract

A comprehensive first-principles study of the energetics, electronic and magnetic properties of Co-doped GaN(0001) thin films are presented and the effect of surface structure on the magnetic coupling between Co atoms is demonstrated. It is found that Co atoms prefer to substitute the surface Ga sites in different growth conditions. In particular, a CoN/GaN interface structure with Co atoms replacing the first Ga layer is preferred under N-rich and moderately Ga-rich conditions, while CoGa_x/GaN interface is found to be energetically stable under extremely Ga-rich conditions. It’s worth noting that the AFM coupling between Co atoms is favorable in clean GaN(0001) surface, but the existence of FM would be expected to occur as Co concentration increased in Ga-bilayer GaN(0001) surface. Our study provides the theoretical understanding for experimental research on Co-doped GaN films and might promise the Co:GaN system potential applications in spin injection devices.

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I. INTRODUCTION

Diluted magnetic semiconductor (DMS) have much attention because of potential applications in spintronic devices\(^1\)\(^-\)\(^4\). In recent years, Co:GaN system is of particularly importance due to its potential application in magneto electronic devices. Experimental and theoretical studies have been reported\(^5\)\(^-\)\(^10\) to investigate magnetic properties in Co:GaN films. A strong ferromagnetic(FM) ordering with \(T_c\) extending \(\sim 250K\) is found by S. Dhara et.al\(^5\) in Co-doped n-GaN by metal-organic chemical vapor deposited(MOCVD). To explore the magnetism origination, W.Kim\(^9\) supposed that the magnetism is origin from the CoGa\(_x\) or Co clusters in Co-doped GaN films. Based on effect of doping concentration, S. Munawar Basha et. al\(^10\) concluded that only lower percentage of cobalt doped GaN is suitable for spintronic application and supposed the decrease in magnetic moment is attributed to the formation of secondary phase or cobalt cluster, which contributes to the antiferromagnetic(AFM) states. Hence, the microscopic nature of magnetism in the Co-doped GaN films has remained unclear.

Moreover, there are many meaningful studies on magnetic characteristics in Co-doped ZnO surfaces\(^11\)\(^-\)\(^14\), which motivate us to explore the magnetism in the Co-doped GaN surfaces. In addition, the structural properties of GaN surfaces depend sensitively on the orientation of the surface termination and reconstructions\(^15\)\(^,\)\(^16\). The GaN(0001) has been demonstrated to have a better surface morphology, and an incommensurate laterally contracted Ga bilayer structure\(^17\) can be existed in extremely Ga-rich conditions, which are typical of molecular-beam epitaxy(MBE) growth. It is believed that the surface structure and morphology is more sensitive to magnetic exchange interaction. Therefore, it is essential to explore the magnetism of Co-doped GaN(0001) under different growth conditions, which is important for the better understanding of the interfacial implications of heterojunction thin films in further experiments.

In this paper, energetics and magnetic properties of Co-doped clean and Ga-bilayer GaN(0001) surfaces are systematically studied by first-principles calculations. It is found that Co atoms show a strong surface sites preference, while different interface structures would be formed in the two surfaces above. The magnetic calculations show that the preferred state of Co-doped GaN(0001) thin film is antiferromagnetic under N-rich conditions, but becomes ferromagnetic with increasing Co concentration under extremely Ga-rich con-
ditions. Finally, electronic structure calculations for the AFM and FM configurations are performed to explain the existence magnetic mechanism in two different GaN(0001) surfaces respectively.

II. COMPUTATIONAL DETAILS

Our calculations are performed using the projected augmented wave (PAW)\textsuperscript{18} method as implemented in the Vienna \textit{ab-initio} simulation package (\textsc{vasp})\textsuperscript{19}. Exchange-correlation energy functional is treated in the Perdew-Burke-Ernzerhof of generalized gradient approximation (GGA-PBE)\textsuperscript{20}. The valence electron configurations for Co, Ga and N are considered as $3d^74s^2$, $3d^{10}4s^24p^1$ and $2s^22p^3$, respectively. The wave functions are expanded in plane wave basis with a kinetic energy cut off of 450 eV. As reported in our previous theoretical studies\textsuperscript{21}, we model the GaN(0001)-$2 \times 2$ surface and the Ga-bilayer GaN(0001)-$\sqrt{3} \times \sqrt{3}$ surface to simulate the different growth conditions. In both two configurations above, the upper three bilayers of GaN and adlayers are allowed to relax, while the bottom bilayer of GaN and the saturating pseudo-H atoms are fixed to mimic bulk substrate. The vacuum region for the clean and Ga-bilayer surface are adopted as 11 Å and 13 Å respectively. Brillouin zones are sampled using $4 \times 4 \times 1$ and $6 \times 6 \times 1$ Monkhorst-Pack grids for the $(2 \times 2)$ and $(\sqrt{3} \times \sqrt{3})$ supercell, respectively. All the atomic positions except the bottom GaN bilayers are relaxed until the force exerted on each active atom was less than 0.03 eV/Å.

III. RESULTS AND DISCUSSIONS

We firstly investigate various configurations (Ga-substitute, N-substitute and the interstitial site) with one Co atom doped in bulk GaN (including 72 atoms), and find that Co atom prefers to locate at the Ga-substitutional site, which is in accordance with the experiment results from S. Dhara\textsuperscript{5}.

A. Energetics of Co-doped GaN(0001) surfaces

Our results for the energetics of Co-doped clean GaN(0001) surface are summarized in Fig. 2 including its Co-coverage dependence up to 1 ML. The different configurations are
FIG. 1. (Color online) (a) (top-view and side-view) Models of Co-doped clean GaN(0001) surface, corresponding to the N-rich conditions or moderately Ga-rich conditions. (b) (top-view and side-view) Models of Co-doped Ga-bilayer GaN(0001) surface, corresponding to the extremely Ga-rich conditions. The vacuum regions are not shown here.

denoted as \((n_1/n_2/n_3)\), where \(n_1\), \(n_2\) and \(n_3\) are the numbers of substituting Co atoms in the first, second and third bilayer, respectively (see Fig. 1(a)).

From Fig. 2 it appears that at 0.25 ML concentration, Co prefers to locate in the first bilayer. The total energy for Co in the second bilayer is about 0.65 eV higher, while the energy further increases by 1.48 eV for Co in the third bilayer. As the Co concentration increased to 0.5 ML, the \((2/0/0)\) configuration becomes more stable than the \((1/1/0)\) with 0.62 eV lower in energy. In addition, in the \((2/0/0)\) configuration, the structure with Co atoms replacing two closest Ga atoms is examined to be the most preferable. As the Co
FIG. 2. (Color online) Relative energies of different configurations for Co-doped the clean GaN(0001) surface. Different configurations are labeled as $(n_1/n_2/n_3)$, $n_1$, $n_2$ and $n_3$ are the Co atoms doped in the first, second, and third GaN bilayer in a $2 \times 2$ supercell.

concentration further increased to 0.75 ML, the most stable configuration still has all Co impurities in the first GaN bilayer, and the configurations $(2/1/0)$ and $(0/3/0)$ are 1.05 eV and 1.63 eV higher in energy, respectively. At 1 ML Co concentration, the configuration $(4/0/0)$ becomes energetically stable. Here we only consider the comparison with configurations of $(2/2/0)$, $(0/4/0)$ and $(0/4/0)$, which is already enough to find the distribution regularity. Therefore, for Co-doped clean GaN(0001) surface, Co atoms prefer to substitute the surface Ga sites.

On the other hand, site preference of Co-doped the Ga-bilayer GaN(0001) surface are investigated in the same way, $(n_1/n_2/n_3/n_4/n_5)$ is used to denote the different incorpo-
FIG. 3. (Color online) Relative energies of different configurations for Co-doped the Ga-bilayer GaN(0001) surface. Different configurations are labeled as \( (n_1/n_2/n_3/n_4/n_5) \), where \( n_1, n_2, n_3, n_4, n_5 \) are the Co impurities incorporated in the outer and inner Ga adlayers, first, second and third GaN bilayer in a \( \sqrt{3} \times \sqrt{3} \) supercell respectively.

At 0.33 ML concentration, Co prefer to substitute the Ga atoms of outermost layer in Ga bilayer\([(1/0/0/0/0)]\), the total energy of this configuration is 1.51 eV lower than that of Co incorporated in the first GaN bilayer\((0/0/1/0/0)\). The optimized structure shows that
Co atom and the underlying Ga atoms are connected tighter, which completely destroy the original flatting Ga bilayer structure. As the Co concentration increased to 0.67 ML, the additional impurity preferentially still occupies at Ga sites in the outermost layer of Ga bilayer\([2/0/0/0/0]\) and more Co atoms bond with the underlying Ga atoms. Same situation continues to maintain as Co concentration further increases to 1 ML, the \((3/0/0/0/0)\) configuration becomes the most stable. As the Co concentration further increases to reach 1.33 ML, the most stable configuration is \((4/0/0/0/0)\). Interestingly noted in the configuration (the optimized structure of \((4/0/0/0/0)\) is shown in Fig. 7(b)), Co atoms replace the outermost Ga layer and connect closely with underlying Ga layer, which makes the original Ga bilayer structure turning to be a cluster-like CoGa bilayer. According the calculation results, the average vertical distance between the newly formed Co layer and the underlying Ga layer is calculated as 1.93 Å, which is significantly reduced compared to the original average distance (2.37 Å) between the outermost Ga layer and underlying Ga layer. The surface characteristic and calculated data demonstrate that a flatting tightly-coupled CoGa bilayer would be formed upon GaN bilayer in this case. Experimentally, CoGa alloys have been observed upon GaN(0001) substrate surface in system of Co deposition on GaN(0001) surface by MBE under excess Ga fluxes\(^7\).

To summarize, the preferred site of Co-doped GaN(0001) surface keeps always on the outermost surface, which is completely different from our previous study on Al incorporation in GaN(0001) surface that Al atoms prefer to migrate into deeper regions\(^21\).

With the relative energies calculated above, the stability of diagram for Co:GaN(0001) can be derived as a function of Ga chemical potential to confirm the growth mechanism. The relative formation energy \(E_{\text{form}}\) can be written as:

\[
E_{\text{form}} = E_{\text{total}} - E_{\text{refer}} - \Delta n_{\text{Co}} \mu_{\text{Co}} - \Delta n_{\text{Ga}} \mu_{\text{Ga}} - \Delta n_{\text{N}} \mu_{\text{N}}
\]

Almost relevant definitions were given in our previous study\(^21\). \(\Delta n_{\text{Co}}\) is the excess or deficit of Co atoms with respect to the reference. A face centered cubic (fcc) crystal structure are adopted for Co-bulk. The \(\mu_{\text{Co}}=\mu_{\text{Co}(\text{bulk})}\) corresponds to the Co-rich conditions. The obtained relative formation energy is a function of \(\mu_{\text{Ga}}\) ranging from \(\mu_{\text{Ga}(\text{bulk})} + \Delta H_{\text{f}}^{\text{GaN}}\) to \(\mu_{\text{Ga}(\text{bulk})}\).

Fig.\(^4\) gives the the calculated formation energies for the most favorable models of various Co concentration in two GaN(0001) surfaces. In case of N-rich conditions, the configuration with lowest energy is \((4/0/0/0)\), meaning that a CoN/GaN interface structure could be formed.
FIG. 4. (Color online) Relative formation energies versus the Ga chemical potential $\mu_{Ga}$ for all possible configurations of Co-doped clean and Ga-bilayer GaN(0001) surfaces. The zero energy as indicated with a horizontal dot line corresponds to the formation energy of the clean GaN(0001) surface.

with the CoN bilayer formed upon the GaN(0001) substrate surface. Same interfacial behavior could also appeared in Fe doping GaN film\(^\text{22}\). In the optimized structures of (4/0/0), the bond length of Co-N is calculated as 1.90Å, which is similar to that of Ga-N as 1.97 Å. The doping of Co atoms doesn’t destroy the original GaN fame with exhibiting a flattening appearance and encourages the bonding of Co atoms with N atoms, which caused the CoN bilayer matching well with GaN(0001) surface structure. Similar fundamental feature of
CoZnO(0001) films growth and structures has been reported in previous experiment study. While under the extremely Ga-rich conditions, the Ga overlayer with an incorporated Co impurity becomes most favorable \([1/0/0/0/0]\), indicating that CoGa_x/GaN interface is thermodynamically stable. According to our calculations, structures with higher Co concentration could be also energetically favorable except the configuration \((4/0/0/0/0)\), the higher formation energy reveals that this surface structure would not be easily formed under natural growth environment, but might be achieved by changing external environment.

B. Magnetic properties of Co-doped GaN(0001) surfaces

After achieving the most stable configuration for different Co concentrations, we investigate the magnetic coupling between the Co atoms in two GaN(0001) surfaces. For Co-doped clean GaN(0001) surface, it is found that the magnetic moment of Co atom is 2.99 \(\mu_B\) in the configuration \((1/0/0)\), which is almost equal to a free Co atom situation \((3 \mu_B)\). Next, the stable \((2/0/0), (3/0/0), (4/0/0)\) configurations are adopted to study the magnetism of Co-doped clean GaN(0001) surface in calculations. Both FM and AFM for each of these configurations are taken into consideration to identify the most preferred magnetic states. Interestingly, the energy differences between FM and AFM \(\Delta E_{FM-AFM}\) are calculated above zero, as shown in Fig. In detail, AFM is found to be more favorable over the FM by higher energy difference\((284\text{ meV})\) in the configuration \((2/0/0)\), which indicates the AFM is relatively stable. For the \((3/0/0)\) and \((4/0/0)\) configurations, the \(\Delta E_{FM-AFM}\) are calculated as 134 and 259 meV respectively, indicating AFM state is still preferred over the FM state. Therefore, it is concluded that the AFM coupling could be stable exist between Co atoms in clean GaN(0001) surface. Our results provide an evidence for the previous experiment study which supposed the AFM state would appear in Ga_{1-x}Co_xN alloys under N-rich conditions. Coincidentally, same AFM phenomenon have been reported experimentally in CoZnO(0001) films.

For Co-doped Ga-bilayer GaN(0001) surface, it is found that one Co atom could induce the magnetic moment of 1.12 \(\mu_B\) into surface system. Then, the \(\Delta E_{FM-AFM}\) of \((2/0/0/0/0), (3/0/0/0/0), (4/0/0/0/0)\) configurations are calculated as shown in Fig. Actually, the \((2/0/0/0/0)\) configuration presents a marginal AFM behavior because of it’s lower energy difference\((\Delta E_{FM-AFM}=23.2\text{ meV})\). But a tendency of FM behavior appears as Co concen-
FIG. 5. (Color online) Energy difference between the FM and AFM configurations as a function of various stable configurations in two cases of GaN(0001) surface. The zero energy difference is indicated with a horizontal line.

In detail, the (3/0/0/0/0) configuration shows FM state with a tiny energy difference ($\Delta E_{FM-AFM} = -4.7$ meV). What’s interesting is that $\Delta E_{FM-AFM}$ of the (4/0/0/0/0) configuration shows a large negative value(-353 meV) with the total magnetic moment of 4.79 $\mu_B$. The calculated results demonstrate that FM coupling interaction will become more effective with increasing Co concentration in Ga-bilayer GaN(0001) surface, especially a relatively stable FM state is found at 1.33 ML concentration. But unfortunately, the higher formation energy of (4/0/0/0/0) configuration is a negative factor for obtaining such a stable FM structure. It is therefore expected that the FM (4/0/0/0/0) configuration might be achieved based on some special experimental tech-
For better understanding the magnetic mechanism in two GaN(0001) surfaces, we choose the (2/0/0) and (4/0/0/0/0) configurations and perform the electronic structure calculations respectively.

The total density of states (DOS) for the (2/0/0) configuration, and the corresponding partial DOS of Co atoms and the partial DOS of neighboring N atoms are plotted in Fig. 6(a1), Fig. 6(a2), Fig. 6(a3), respectively. We note that the total DOS for spin-up and spin-down are identical, therefore there is no net magnetic moment in this case, as shown in Fig. 6(a1). The magnetic moment on each one Co atom (refers to the marked 1Co or 2Co in Fig. 6(a2)) is 1.84 $\mu_B$ and almost comes from the Co-3d orbital (1.77 $\mu_B$). Small contributions
FIG. 7. (Color online) (a) DOS and partial DOS of the configuration (4/0/0/0/0). (b) Iso-surface plots (0.10 e/Å$^3$) for charge density difference of configuration (4/0/0/0/0). (c) Iso-surface plots (0.007 e/Å$^3$) for spin-up and spin-down charge density of FM configuration (4/0/0/0/0).

to the moment derive from the Co-4p(0.06 $\mu_B$) due to the p and d hybridization, see Fig. 6(a$_2$). For each Co atom, the neighboring N atom is polarized non-reversing with a magnetic moment of 0.04 $\mu_B$ which mainly comes from N 2p orbital(0.03 $\mu_B$), as shown in Fig. 6(a$_3$). While for each pair of Co and N atoms, the other is arranged antiferromagnetically observed from the spin-density distribution (Fig. 6(c)), and thus lead to the AFM state in the whole system. Then the charge density difference of (2/0/0/0) is plotted in Fig. 6(b) and shows that there is an obvious charge transfer between Co atoms and Ga atoms around in real space, meanwhile an interaction between Co and neighbored N atoms are observed. The interaction among the three types of atoms (Co, Ga and N atoms) gives an evidence to the formation of Ga$_{1-x}$Co$_x$N in the Co-implanted GaN films under N-rich conditions$^{23}$.

The total DOS for spin-up and spin-down electrons states corresponding to the (4/0/0/0/0) configuration is plotted in Fig. 7(a$_1$). The partial DOS of Co and neighboring Ga atoms
are plotted in Fig. 7(a2) and Fig. 7(a3), respectively. The total spin DOS shows that
the spin-up and spin-down are not symmetric near $E_F$ with exhibited 100% polarization,
therefore shows a half-metallic behavior and there is a net magnetic moment (4.79 $\mu_B$) in
system. The magnetic moment on each Co atom is 1.2 $\mu_B$ and mainly comes from Co-3d
orbital (1.13 $\mu_B$), the remaining contribution to the moment arise from Co-4p, as shown in
Fig. 7(a2). Actually, the electronic states of Co atoms and Ga atoms below contribute to
the half-metallic behavior near the $E_F$ more due to the p-d(Ga-4p and Co-3d) hybridization,
see Fig. 7(a2) and Fig. 7(a3), respectively. Inversely, without the polarization from N atom,
the magnetic moments of four Co atoms are in parallel which leading to FM state in this
case (see Fig. 7(c)), it’s totally different with configuration (2/0/0). Combined with the
analysis of optimized geometric structure of (4/0/0/0/0) in Sec II.A, thus, it is believed
that the formation of tightly-coupled CoGa bilayer upon the GaN(0001) surface is directly
benefit to FM ordering. The charge density difference in real place (Fig. 7(b)) shows that
there is an obviously charge transfer phenomenon from Co atoms to the underlying Ga
atoms, even drives the activity of Ga atoms of the first GaN bilayer. This provides an
intuitive understanding for the formation of CoGa alloys upon GaN(0001) substrate surface
under excess Ga fluxes in experiment study\(^2\).

IV. CONCLUSIONS

In summary, we have carried out a detail DFT study for the energetics and magnetic prop-
erties of Co-doped GaN(0001) thin films. Our results on the Co:GaN(0001) system suggest
that it is more favorable for Co to substitute Ga atoms of the outermost Ga layer near
surface than inside the bulk. It shows that CoN/GaN interface could be easily formed under
N-rich conditions, while under Ga-rich conditions, CoGa\(_x\)/GaN interface could be energeti-
cally favorable. Moreover, it is demonstrated for Co atoms to coupling antiferromagnetically
in clean GaN(0001) surface, and the AFM coupling is not sensitive to doped concentration.
But to the case of Co doping in Ga-bilayer GaN(0001) surface, it is possible that the coupling
between Co atoms become ferromagnetically as the Co concentration increased, especially
at 1.33 ML concentration. It is concluded that such magnetism in Co:GaN(0001) thin films
is directly related to experimental growth conditions and the Co:GaN system is expected to
have potential applications in the field of spin injection devices.
V. ACKNOWLEDGMENTS

The authors thank Prof. Xu Zuo of NKU for helpful discussions and acknowledge the computing facilities of the Center for Computing of Tsinghua University. This work was supported by National Natural Science Foundation of China (Nos.51062003,61264005), Young Scientist Program of Jiangxi Province (20122BCB23030), Science Foundation of Jiangxi Province(Nos.GJJ14587, 20133BBE50001) and Innovation Team in JXSTNU (2013CXTD001).

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