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Magnetic Properties of NiO and MnO by LSDA+U

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***Abstract*—**The spin (m_s) and orbital (m_o) magnetic moment of the antiferromagnetic NiO and MnO have been studied in the local spin density approximation (LSDA+U) within full potential linear muffin-tin orbital (FP-LMTO method with in the coulomb interaction *U* varying from 0 to 10eV, exchange interaction *J*, from 0 to 1.0eV, and volume compression *VC* in range of 0 to 80%. Our calculated results shown that the spin magnetic moments and the orbital magnetic moments increase linearly with increasing *U* and *J*. While the interesting behaviour appears when volume compression is greater than 70% for NiO and 50% for MnO at which m_s collapses. Further increase of volume compression to be at 80% leads to the disappearance of both magnetic moments.

***Keywords*—**spin-orbital magnetic moment, Coulomb interaction *U* and exchange interaction *J*, volume compression *VC*, LSDA+U.

I. Introduction

FOR over a decades, many researchers interested in the electronic structures and the magnetic properties of transition metal monoxides (TMO), such as MnO, FeO, CoO and NiO, have been widely investigated both experimentally and theoretically. Among these compounds, NiO and MnO are one of the most potentially applicable compound being used in variety of applications for instance high-temperature super-conductors [1], and electrochromic devices [2]. It is widely accepted that NiO and MnO are a type II antiferromagnetic insulator, the magnetic moments are aligned ferromagnetically on the (111) plane and the Neel temperature (T_N) of 523K and 122K, respectively.[3],[4]. The number of working experimental researchs have been recently dedicated to discriminate its relevant physical and magnetic properties including Coulomb interaction *U* [5], the magnetic moments [6]-[8] and lattice parameter [9]Previous experiments gave the total magnetic moment of NiO is equal to $1.77\mu_B$[7], $1.64\mu_B$ [6] and MnO is $4.58\mu_B$ [10]. Meanwhile, theoretical studies on NiO and MnO have been accordingly carried out by various calculation methods including the local spin density approximation plus Coulomb interaction *U* (LSDA+U) [11],[12], the local spin density approximation plus dynamical mean-fields theory (LSDA+ DMFT) [13], the self-interaction correction local spin density approximation (SIC-LDA)[14], [15], SIC+LDA+U [16], [17], Constrained LDA [18], and so on. Although the electronic and magnetic properties of NiO and MnO have been studied within LSDA+U method by many pioneered works, but the effect of the Coulomb interaction *U*, the exchange interaction *J* and the volume compression on both spin and orbital magnetic moment have not yet been investigated.

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Thereby, in this work, we report on the pioneered study focusing on the effect of the parameter *U, J* and the *VC* on spin and orbital magnetic moment of NiO and MnO by using the LSDA+U+FP-LMTO method. The calculated results are described guiding to the better knowledge on magnetic phenomenon and properties of this compounds.

II. Calculation Details

In this work, the magnetic moments of NiO and MnO were calculated by using the local spin density approximation plus the Coulomb interaction (LSDA+U) [12] method within the full potential linear muffin-tin orbital (FP-LMTO) [19]. To determine the orbital magnetic moment, the spin-orbital coupling is simultaneously included in the self-consistent loop. The LSDA+U Hamiltonian is given by [12]

$$\hat{H}_{LSDA+U} = \hat{H}_{LSDA} + \hat{H}_U - \hat{H}_{Dc} \tag{1}$$

where $\hat{H}_{LSDA}$ and $\hat{H}_{Dc}$ represent LSDA Hamiltonian and double counting correction for $\hat{H}_U$, respectively. With the Coulomb interaction U and exchange interaction J parameters, one can write $\hat{H}_{Dc}$ and $\hat{H}_U$ as [12]

$$\hat{H}_{Dc} = \frac{1}{2}UN(N-1) - \frac{1}{2}J\sum_{\sigma} N^{\sigma}(N^{\sigma}-1) \tag{2}$$

$$\begin{aligned}\hat{H}_U = {} & \frac{1}{2}\sum_{\{m\},\sigma} V(mm';m''m''')n^{\sigma}_{mm''}n^{-\sigma}_{m'm'''} \\ & + \frac{1}{2}\sum_{\{m\},\sigma}\left[V(mm';m''m''')\right. \\ & \left. -V(mm';m'''m'')\right]n^{\sigma}_{mm''}n^{-\sigma}_{m'm'''}\end{aligned} \tag{3}$$

When $N^{\sigma} = Tr(n^{\sigma}_{mm'})$ and $n^{\sigma}_{mm'}$ is the d occupation number matrix of spin σ. Meanwhile the relation between screen Coulomb interaction $V(mm';m''m''')$ and Slater integral F^K is given by

$$V(mm';m'''m'') = \sum_{k=0}^{2l} c^K(lm,lm'')c^K(lm',lm''')F^K \tag{4}$$

where $c^K(lm,lm'')$ is a Gaunt coefficient. For NiO, MnO all 3d TMO materials, three Slater integrals of F^0, F^2 and F^4 are involved in calculation, which they connected to parameter U and J by $U = F^0$ and $J = (F^2 + F^4)/14$ [13].

Our calculations assumed NiO and MnO as a type II antiferromagnetic with a lattice constant of 7.926 a.u. and 8.393 a.u., respectively. The muffin-tin radii of Ni^{2+}, O^{2-} and Mn^{2+}, O^{2-} ions are 2.179, 1.783 a.u, and 2.346, 1.843 a.u., respectively and the volume compression was defined by *V/V0* ratio. The effect of *U, J* and *VC* on the magnetic properties of

NiO and MnO were studied varying *U*, *J* and *VC* ranging from 0-10 eV, 0-1.0eV and 0-80% , respectively.

III. Results And Discussions

A. Effect of U and J

Magnetic moments of NiO and MnO were evaluated as function of Coulomb interaction U and exchange interaction J, which the corresponding results are illustrated in Figure 1 and Figure 2, respectively. The calculation was carried out by the LSDA+U method at designed *J* of 0.40eV, 0.60eV, 0.80eV and 0.95eV and normal volume without compression. The result shows that total magnetic moments of NiO and MnO gradually increase with increasing Coulomb interaction as shown in Figure 1. This feature is understandable in view of the reason that the Coulomb interaction *U* enhances the electron localization, which leads to magnetic moment gain. Moreover, the 10% change of *U* can induce the change of 0.3% in total magnetic moments. Calculated magnetic moments of NiO and MnO are summarized in Table. 1 in comparison with experimental data and previous results by other calculated methods. Our calculated results reveal that the orbital magnetic moments are underestimated comparing to the experiment results [21] meanwhile the spin magnetic moment and Coulomb *U* values are agreement with experimental data [5],[7]

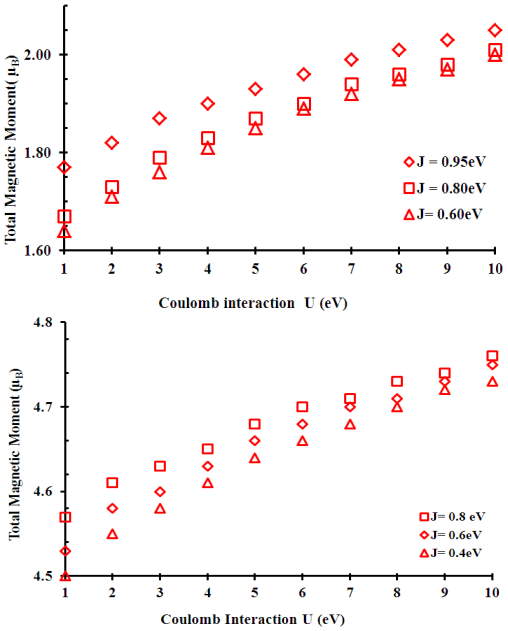

Fig. 1 The total magnetic moment at normal pressure of NiO(top) and MnO(bottom) depend on the Coulomb interaction *U* for all *J*

Furthermore, In Figure 2 exhibits the total magnetic moment of NiO and MnO also strongly depend on *J* for all *U* . The calculated results of total magnetic moments are proportional to the exchange interaction *J*, which the ratio of m_t and *J* approximated 1:1. Since on the atomic scale, the exchange interaction J tends to align neighbour spins so the total magnetic moment increase with increasing of *J*. Therefore we can conclude that the magnetic moment depend on both U and *J*. It is noticed that total magnetic moments of NiO and MnO increase with increasing of *U* and *J*.

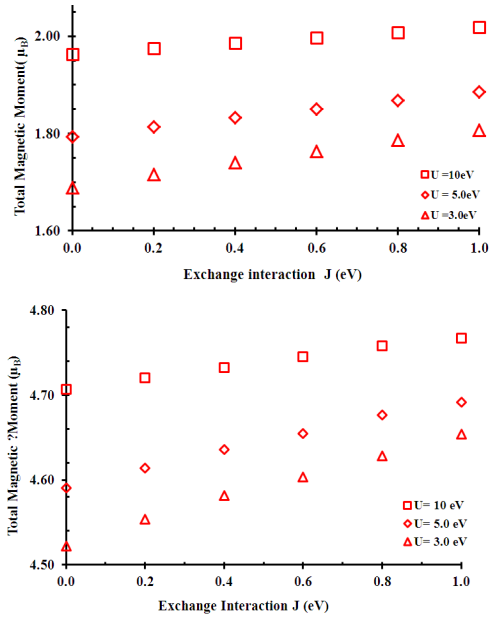

Fig. 2 The total magnetic moment at normal pressure of NiO(top) and MnO (bottom) depend on the Coulomb interaction *J* for all *U*.

B. Effect of VC

Figure 3 shows the variations of the spin (m_s) and orbital (m_o) magnetic moments of NiO and MnO as a function of volume compression at *U* of 8.0eV. It is noticed that m_s and m_o decrease with increasing *VC* whereas the spin magnetic moment slightly decrease. At the compression range of 0-60% for NiO and 0-50% for MnO, the m_s decreases from its original value by 0.6% (NiO) and 1.2% (MnO) as the *VC* increases by 10%. As the *VC* is larger than 70%, the spin magnetic moment of NiO and MnO rapidly decreases to zero. Meanwhile, the value of orbital magnetic moment of NiO gradually decreases by 2.0% for every 10% increasing *VC* until the collapse of m_o occurs at 80% of *VC*. This strange behaviour may be originated from the delocalization of most electrons.Furthermore, Figure 3 shown the interesting phenomena while volume was compressed 60%. The values of spin and orbital magnetic moment of NiO equate MnO. In our notion, at this point, the electronic structure of NiO and MnO should be similarly. This strange phenomena is quite interesting and still in doubt therefore the further deep researches not only experimental but also theoretical works are required for better clarification

TABLE I
CALCULATED RESULTS AND EXPERIMENTAL DATA OF THE COULOMB INTERACTION *U* (EV), THE EXCHANGE INTERACTION *J* (EV), THE SPIN M_S (μ_B) AND THE ORBITAL M_O (μ_B) MAGNETIC MOMENT OF NIO AND MNO AT NORMAL PRESSURE

Parameter	NiO				MnO		
	Our Work	GGA[11]	LSDA+*U* [12]	Experiment	Our work	LDA+*U* [17]	experiment
U	1.0-10	6.20	8.00	5.80 [5]	1.0-10	6.9	-
J	0-1.0	1.36	0.89	-	0-1.0	0.86	-
m_s	1.39-1.85	2.00	1.64	1.64 [6] , 1.77[7], 1.90 [8],[21]	4.42-4.77	4.59	4.58[10] 4.79[22]
m_o	0.14-0.17	-	0.29	0.32[21]	-		-

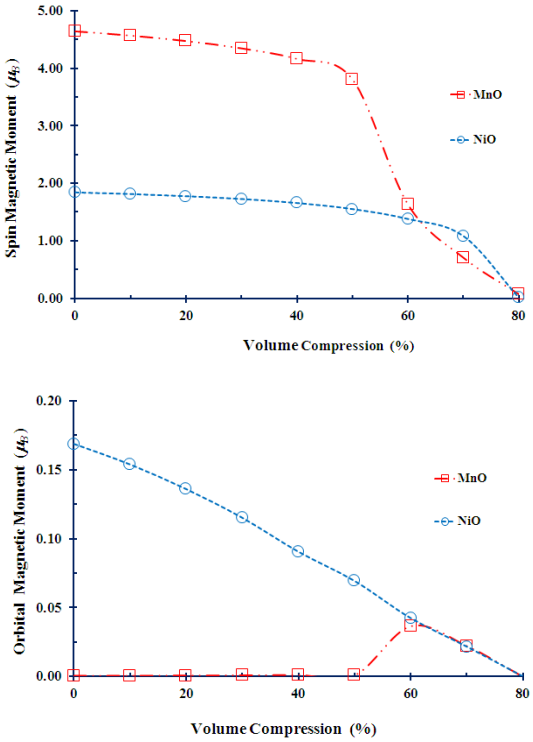

Fig. 3 Spin (top) and orbital (bottom) magnetic moment of NiO (circle) and MnO (rectangular) depend on the VC

IV. CONCLUSION

Effect of the Coulomb interaction U, exchange interaction *J* and volume compression *VC* on magnetic properties of NiO and MnO were studied by using the local spin density approximation plus the Coulomb interaction method within the full potential linear muffin-tin orbital. Our results revealed that the total magnetic moments of NiO and MnO depend on Coulomb interaction *U* , exchange interaction *J*, and volume compression *VC*. The magnetic moment increase with increasing of Coulomb and exchange interaction, but decrease with increasing volume compression. Moreover, at rather compression, The *VC* exceed 80% causing the disappearance of magnetic moment.

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REFERENCES

[1] M. Rubinstein, R.H. Kodama, S.A. Makhlouf, J Magn Magn Mater. 2234, 289 (2001).
[2] H. Liu, W. Zheng, X. Yan, and B. Feng, J. Alloy Compd. 462, 356(2008).
[3] C.G. Shull, W.A. Strauser, and E.O. Wollan, J. Phys. Rev. 83, 333 (1951).
[4] J. Baruchel, M. Schlenker, K. Kurosawa, and S. Saito, J. Philos. Mag. B 43, 853 (1981).
[5] J.C. Fuggle, G.A. Sawatzky, J.W. Allen, B. Brandow, Narrow Band Phenomena, (Plenum, New York, 1988).
[6] H. A. Alperin, J. Phys. Soc. Jpn. Suppl. B. 17,12 (1962).
[7] I. Barin, Thermochemical Data of Pure Substance, (VCH-Cambrige,1989).
[8] B.E.F. Fender, A.J. Jacobson, F.A. Wedgewood, J. Chem. Phys. 48, 990 (1968).
[9] R. W. G. Wyckoff, Crystal Structure, Interscience, New York, 1964.
[10] A.K. Cheetham and D.A. Hope, J. Phys. Rev B, 27, pp 6964-6967, 1983.
[11] Tuo Cai, Huilei Han , You Yu , Tao GaO, Jiguang Du and Liangghuan Hao, J. Physica B 404, 89 (2009).
[12] S.K. Kwon and B.I. Min, J. Phys. Rev. B 62, 73(2000)
[13] O. Miura and T Fujiwara, J. Phys. Rev. B 77, 195124 (2008).
[14] A. Svane Du and O. Gunnarsson, J. Lett.65,vol 9, pp 1148-1151, 1990.
[15] Z. Szotek, and .W.M. Temmerman, J. Phys. Rev.B.47, vol 7, 4029-4032, 1992.
[16] V.I. Anisimov, J Zaanen and O.K Anderson, J. Phys. Rev. B, 44, vol 3, pp 943-954, 1991.
[17] D.W. Boukhvalov, A.I. Lichtenstein and V.I. Anisimov, J Phys. Rev. B.65,pp 184435-1- 1184435-6, 2002.
[18] I.A. Nekrasov, M.A. Korotin and V.I. Anisimov, "cond-mat/0009107v1.
[19] S.Y. Savasov, J. Phys. Rev. B54, 16470–16486 (1996).
[20] F. M. F. de Groot, J. C. Fuggle, B. T. Thole, and G. A. Sawatzky, J. Phys. Rev. B 42 , 5459 (1990).
[21] V. Fernandez, C. Vettier, F. de Bergevin, C. Giles, and W. Neubeck, J. Phys. Rev. B 57, 7870(1998).
[22] F.P. Koffyber and F.A. Benko, J Appl. Phys. 53, 1173(1982.