Oxygen vacancies of YIG influence on the MPE in Au/YIG heterostructures

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Abstract. The oxygen vacancies of $Y_3Fe_5O_{12}$(YIG) influence on the magnetic proximity effect (MPE) in Au/YIG heterostructures has been studied by first principles calculations based on the density functional theory (DFT). The results show that one oxygen vacancies can reduce the two neighbor Fe^{3+} to Fe^{2+} and MPE in Au/YIG is strongly depended on the valence states of Fe ion. The induced magnetism in Au atoms is found to increase with the increasing concentration of Fe^{2+}. In oxygen vacancy free case (Fe^{3+} state), weak electron states hybridization of Fe(3d) and Au(5d) is found form the projected electron density of states distribution, while after introduce the oxygen vacancy (Fe^{2+} state), the states of Fe(3d) are generally move to higher energy states and hybridize with the Au(5d) states. Moreover, more states of Fe^{2+} at tetrahedral sites are hybridize with the Au(5d) states compare with that of Fe^{2+} at octahedral sites, therefore a net magnetic moment induced Au atoms is found and the direction of spin polarization of Au atoms is paralleled to Fe ion at tetrahedral sites.

1. Introduction

In recent years there has been a very strong interest in the magnetic proximity effect (MPE) in Au(Pt)/$Y_3Fe_5O_{12}$ (YIG) heterostructures, which directly affects the interpretations of a number of recent important experimental observations of spintronics in the Au(Pt)/YIG system [1-3]. The magnetic proximity effect is a phenomenon of ferromagnetic ordering of spins within several atomic layers of a noble metal (NM) film in close proximity to a ferromagnetic material [4-6]. In previous both theoretical and experimental study, the MPE in Au/YIG is insignificant which is an order of magnitude weaker than that in Pt/YIG [7-9]. However, Francesco Pineider et al. found that the MPE in Au/FeO is much stronger than that in Au/Fe_2O_3 [10], which indicated MPE maybe existed in the Au/Iron oxide heterostructures while the Fe^{3+} is reduced to Fe^{2+}. In each YIG unit cell, one oxygen vacancy can reduce two Fe^{3+} to Fe^{2+} at octahedral and tetrahedral sites, respectively [11]. With the aim of providing a theoretical prediction the MPE in Au/YIG bi-layered system while oxygen vacancies induced in YIG layer, first-principles calculations were carried out. The calculations are based on density functional theory (DFT). The results show that with the increasing of oxygen vacancies concentration, the MPE in Au/YIG become stronger and stronger because of the Au(5d)-Fe^{2+}(3d) states hybridization.

2. Calculation method

In this paper, we applied the first-principles calculation method based on the density functional theory (DFT) to study oxygen vacancies of YIG influence on the MPE in Au/YIG heterostructures. The Perdew-Burke-Ernzerhof (PBE) exchange-correlation function [12] for the

generalized-gradient-approximation (GGA) and a plane-wave basis set within the framework of the projector augmented wave (PAW) method were employed [13]. For the sampling of the Brillouin zone, we employed the Monkhorst-Pack scheme and used 3×3×1 k-point grids for calculations. The DFT+U formalism developed by Dudarev *et al* was used to account for the strong on-site Coulomb repulsion for the localized Fe (*3d*) states. The value of "U" used for Fe (3d) states in our calculations is 4.3 eV. This value has been widely used in YIG with slight Ce doping and other systems containing Fe^{3+} cations [14]. To model the Au/YIG interfaces, we constructed a superlattice structure with a slab of YIG(100) of about 6 angstrom thick along with four Au(100) atomic layers on top. A 15 angstrom thick vacuum layer was inserted to avoid the interaction between the two cells as shown in Figs. 1(a) and 1(b). During the relaxation process, the in-plane lattice constant was fixed at the experimental value of the bulk YIG material, with the dimensions of 12.376×12.376 $Å^2$. Due to the lattice misfit, the Pt layers were under the biaxial tensile strains of 1.1%. All atoms were fully relaxed until the calculated force on each atom is smaller than 0.02 eV/Å.

3. Results and Discussions

In a unit cell of YIG, the octahedral Fe (Fe^{oct}) arranges in 8 body-centered cubic (BCC) subunit cells; the yttrium and tetrahedral Fe (Fe^{tet}) are located on each surface of the 8 subunit cells, and the oxygen ions surrounds the Y and Fe ions with dodecahedral, octahedral, and tetrahedral coordination respectively. Due to the super exchange effect, the spin polarization directions of Fe^{tet} and Fe^{oct} are opposite. For convenient, we denote spin up and spin down directions for the Fe^{tet} and Fe^{oct} ion, respectively. The calculations considered 6-Å-thick YIG slabs because the induced moments converge at a YIG slab thickness of 6 Å when the thickness was increased from 3 Å to 9 Å. To investigate the effects of the oxygen vacancies of YIG on the MPE in Au/YIG, one, two, and four oxygen vacancies are induced at the interface of Au/YIG structure, respectively, and a 15 angstrom thick vacuum layer was inserted to avoid the interaction between the two cells as shown in fig. 1(a)-(c). Fig. 1(e) shows the calculated moments (μ_B/Au atom) in each Au layer. One can see that by inducing oxygen vacancies (VO) at Au/YIG interface, the magnetic moments of first Au atomic layer significantly enhanced comparing with the defect free case and then rapidly decreased. The average induced magnetic moment over all Au atoms, which indicates the strength of the MPE, are $0.006\mu_B$/atom, $0.008\mu_B$/atom, $0.015\mu_B$/atom for 1VO, 2VO, and 4VO cases. For comparing, the Pt/YIG system without defects is also calculated, and the average induced magnetic moment over all Pt atoms is 0.070, which is still much larger than Au/YIG(4VO) system. Fig.1 (d) shows the structure of Au/YIG(2VO) system, where the 2VO located at the different atomic layers in the YIG. From the calculated moments in Au as shown in fig.1 (f), it is found that the induced magnetic moments of Au in the Au/YIG(2VO)-Ⅰ system are lager than that in Au/YIG(2VO)-Ⅱ system, which indicates the oxygen vacancies with different distance from the interface have unequal effect on the MPE, and the effect becomes weaker while the oxygen vacancy gradually away from the interface. Table 1 lists out the quantitative results of the magnetic moments induced in different systems. One can see by introducing oxygen vacancies to the YIG lattice, the magnetic moments of Au can enhance several times.

Table 1. In different Au/YIG system as shown in fig.1(a-d), the concentration of Fe^{2+} in YIG layer, the average magnetic moments in 1^{st}-4^{th} Au(Pt) atomic layers (M_1-M_4) and over all Au atoms (M_0) are listed. ΔM is the percent variations in M0 comparing with the oxygen vacancy free case.

System	Fe^{2+}%	M_1	M_2	M_3	M_4	M_0	ΔM
Au/YIG	0%	0.004	0.002	0.002	0.000	0.002	...
Au/YIG(1VO)	7%	0.017	0.006	0.002	-0.001	0.006	200%
Au/YIG(2VO-Ⅰ)	14%	0.022	0.009	0.002	-0.002	0.008	300%
Au/YIG(2VO-Ⅱ)	14%	0.018	0.003	0.000	-0.001	0.005	250%
Au/YIG (4VO)	28%	0.031	0.010	0.013	0.008	0.015	650%
Pt/YIG	0%	0.041	0.065	0.091	0.081	0.070	...

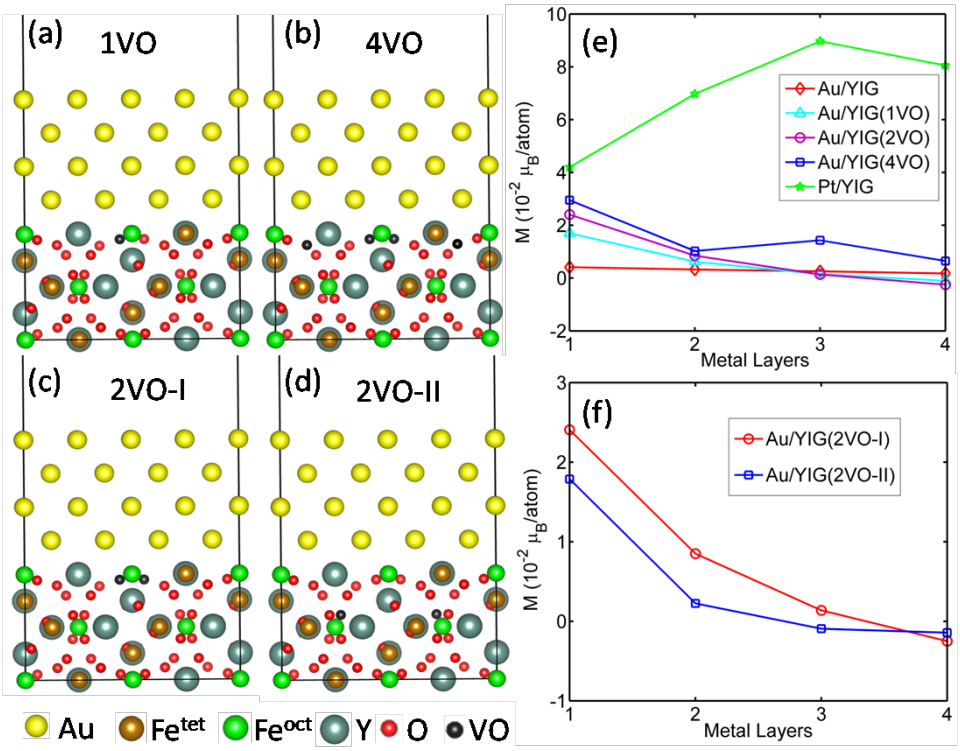

Figure 1. Side view of the calculated crystalline structures of Au(100)/YIG(100) in different oxygen vacancies induced cases: one (a), two(c) and four(b) oxygen vacancies located at the interface; two oxygen vacancies located at the deeper layer of YIG (d). (e)(f) show the induced magnetic moments of each layer Au(Pt) in different oxygen vacancies doped cases.

To reveal the origin of the oxygen vacancies increasing the magnetic moments of Au in Au/YIG system, the projected density of states (PDOS) is studied. Fig. 2(a) and (b) show the PDOS of the interface Au, Fe^{tet} and Fe^{oct} ion in Au/YIG without and with oxygen vacancies cases, respectively. The DOS peaks of Fe^{tet} and Fe^{oct} neighbor to the oxygen ion below the Fermi leave are both located above -7eV, which is agree with other calculation results of PDOS in bulk YIG [11]. On the contrary, the locations of peaks of Fe^{tet} and Fe^{oct} the oxygen vacancy moved 1.56eV and 1.03eV near to the Fermi level, and more states of Fe^{tet} are hybridization with Au 5d states comparing to that of Fe^{oct}. Simultaneously, we can see an additional down spin and up spin peak are added for Fe^{tet} and Fe^{oct}, which indicates the Fe^{3+} is reduced to Fe^{2+} due to the oxygen vacancy. Therefore, through electrons exchange effect between Fe^{2+} and Au, the magnetic moment is induced in Au and the direction is along the spin polarization direction of Fe^{tet}. The different strength of the electron hybridization between Pt and Fe ions induces significantly different magnetic properties of Pt due to direct exchange, which leads to different MPE responses in Pt/YIG with different interface structures.

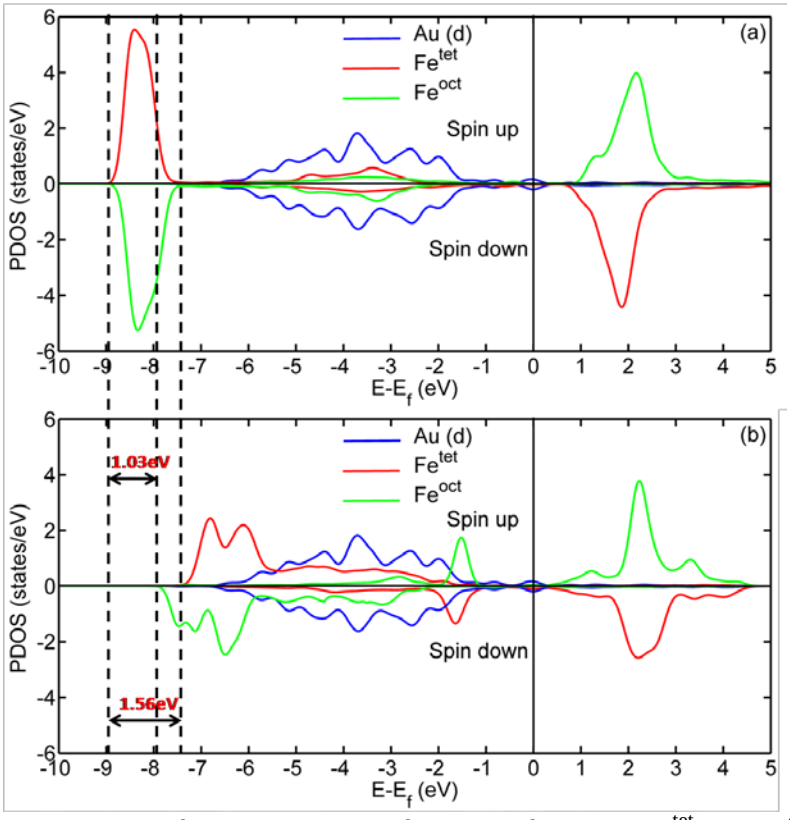

Figure 2. The projected density of states (PDOS) of the interface Au, Fe^{tet} and Fe^{oct} ion in Au/YIG heterostructures without (a) and with (b) oxygen vacancies.

4. Summary

In summary, we studied the oxygen vacancies influence on the magnetic proximity effect of Au/YIG bi-layered heterostructures using first principles calculations. The results show with the increasing concentration of oxygen vacancies at the Au/YIG interface, the MPE in Au/YIG heterostructure become stronger and stronger. The physical origin of this effect can be explained by the projected electron density of states distribution, which can be described as the following: by introducing the oxygen vacancy, the Fe^{3+} in YIG is reduced to Fe^{2+}, then the Fe(3d) states move to higher energy level and hybridize with Au(5d) states. Through the electron exchange effect, magnetism is induced to Au atoms. In addition, we also found he magnetism of Au atom become weaker while the oxygen vacancies are moved further layer from the Au/YIG interface.

Acknowledgments

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