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Strain control of magnetocrystalline anisotropy and energy product of MnGa alloys

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ABSTRACT

We investigate the energy product of MnGa alloys as function of Mn concentration and applied elastic strain. Using the density functional theory (DFT) based method we calculated the magnetocrystalline anisotropy (MAE) and magnetization of Mn–Ga alloys as function of composition, e.g. Mn and Ga, and examined their variation under applied strain. Our calculations show that MAE is very large ~22–27 M erg/cm^3 in all three considered compositions, e.g. MnGa, Mn_3Ga and $Mn_{1.66}Ga$. We show that MAE is very robust in MnGa system and remains large in wide range of concentrations and strains both compressive and tensile. We find that bi-axial tensile strain increases MAE in $Mn_{1.66}Ga$ alloys. Our study shows that the variation of MAE as function of Mn content is related to the change in electronic structure and, specifically, the Fermi level position with electron population variation. We estimated the theoretical limit of the energy product $(BH)_{max}$ of MnGa, Mn_3Ga and $Mn_{1.66}Ga$ alloys as 23.65, 4.06 and 13.64 MGOe, respectively. We find that volume expansion of the MnGa alloys (by appropriate doping) should increase the magnetization and the energy product of these alloys.

1. Introduction

High coercivity magnetic materials exhibiting a perpendicular magnetic anisotropy (PMA) and large magnetic energy product have great application potential in ultrahigh-density perpendicular magnetic recording media, high-performance permanent magnets and spintronic applications such as magnetic tunnel junctions (MTJs) and spin injectors [1–12]. Materials with large PMA possess interesting physical phenomena like efficient spin injection into semiconductors from ferromagnetic contact, giant anomalous Hall effect, and long-lived ultrafast spin procession [13–15]. It is important to explore methods to control the magnetic anisotropy in crystalline thin films to develop functional and highly reliable devices [10,16].

Mn–Ga alloys are PMA materials that have the thermodynamic stability, relatively large spin polarization, low saturation magnetization, square-like magnetization hysteresis loops, and perpendicular anisotropy [17–21]. Mn–Ga binary alloys show strong magnetism and large uniaxial magnetic anisotropy even though these alloys do not contain any noble, rare-earth metals or magnetic elements. Mn_3Ga crystallizes in the $D0_{22}$ structure exhibits a unique combination of low magnetization, high uniaxial anisotropy, high Curie temperature and high spin polarization, which suit the requirements for spin torque memories down to 10 nm in size [19,22]. The structural, magnetic, and transport properties for off-stoichiometric $L1_0$ Mn–Ga films have been investigated in view of their magneto-optical and spintronics applications [12,14,17,18]. $L1_0$- $Mn_{50}Ga_50$ homogeneous films were theoretically predicted to have large magnetocrystalline anisotropy (MAE) of 26 Merg cm^{-3}, moderate magnetization of 2.51 μ_B/Mn, and large theoretical limit of magnetic energy product of 28.2 MG Oe [20,23–25].

Alloys of Mn_xGa_{1-x} with $L1_0$ structure are thermodynamically stable for (x=0.5–0.65) with large MAE [26] $D0_{22}$ structures of Mn_xGa_{1-x} can be stabilized using certain preparation procedures at x=0.5-0.65. It exhibits strong ferrimagnetism, as confirmed by neutron scattering [27]. A high Curie temperature of up to –800 K, large magnetic anisotropy, and composition-sensitive magnetization were also reported in $D0_{22}$-phase polycrystalline bulk samples [28]. Furthermore, Mn_xGa ($1<x<1.8$) alloys with $L1_0$ structures have increasing attention because of potential applications in ultrahigh-density magnetic recording media, permanent magnets and spintronics. Recently, $L1_0$-$Mn_{1.5}Ga$ epitaxial films grown on GaAs (001) have tunable perpendicular coercive field (H_c) from 8.1 to 42.8 kOe, MAE up to 21.7 Merg cm^{-3}, magnetic energy product up to 2.60 MG Oe, controllable magnetization from 27.3 to 270.5 emu cm^{-3} [29].These films are expected to be multifunctional and cost-effective alternative for perpendicular magnetic recording bits and a variety of novel devices with high magnetic-noise resistance and thermal stability.

Previous experimental results have shown that the Mn–Ga

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alloys grown on substrate or in superlattices have affected the tetragonality of the crystal structures, which is attributed to the lattice strain, and this has influence on the magnetic properties and the magnetocrystalline anisotropy of the alloys [18,30–36]. It was found that the crystalline orientation of the Ferromagnetic $L1_0$-MnGa thin films, which epitaxially grown on GaN, sapphire, and MgO substrates, differ due to the influence of the substrate [32]. The in-plane and out-of-plane anisotropy is directly determined by the crystal orientation of the film and could be controlled via selection of the substrates. Moreover, it was reported that the substrate modifies the magnetic properties of $L1_0$-Mn_xGa ($1.2 < x < 1.5$) films. Particularly, the Mn_xGa film is a hard ferrimagnet when grown on GaSb (111), becomes *a* soft ferrimagnet when grown on Al_2O_3 (0001), and exhibits an absence of a net magnetic moment when stabilized on *a* GaSb (100) substrate [33]. These results may be helpful in modifying magnetic anisotropy in thin films for spintronic applications. Therefore, understanding the effect of strain in the magnetocrystalline anisotropy and magnetization of MnGa alloys helps to realize the potential of Mn–Ga alloys for use in various applications.

In this work we performed density functional theory (DFT) electronic structure calculations to examine the magnetocrystalline anisotropy energy (MAE) and magnetization of Mn–Ga alloys as function of compositions (Mn and Ga) and strain. We investigated the structural and magnetic properties of the alloys as function of compositions and under applied strain. Our calculations show that MAE of Mn–Ga alloys are large. We show that MAE in MnGa and Mn_3Ga alloys remains large when moderate bi-axial strain is applied. The tensile strain increases MAE in $Mn_{1.66}Ga$ alloys. The changes in the electronic structure, specifically the Fermi level position with electron population variation, explain the variation of MAE for different Mn content. Our study shows that volume expansion of the MnGa alloys increase the energy product of these alloys.

2. Computational methods

Self-consistent electronic structure calculations were performed for MnGa, Mn_3Ga and $Mn_{1.66}Ga$ alloys. The calculations were carried out using the density-functional theory (DFT) method [37,38] as implemented in the Vienna ab initio simulation package VASP [39]. Projector augmented wave PAW pseudopotentials were used [40]. The generalized gradient approximation (GGA) of Perdew–Burke–Ernzerhof (PBE) form [41] is used for the exchange–correlation functional. We used a $16 \times 16 \times 16$ *k*-points sampling for MnGa and $12 \times 12 \times 9$ for Mn_3Ga and $Mn_{1.66}Ga$, the Blöchl's tetrahedron integration method was used [42]. We set the plane-wave-cut-off energy to 350 eV and we choose the convergence criteria for energy of 10^{-6} eV. Calculations were performed with relaxation of both atomic positions and lattice parameters. We relax the atomic positions of all atoms in the unit cells using Hellmann–Faynmann scheme till forces were less than 0.003 eV/Å. We included the spin–orbit interaction in our calculations and employed force theorem to calculate the $MAE = E(\|) - E(\perp)$ as difference of energies of the system with magnetization along and perpendicular to the easy axis [23,43,44].

3. Results and discussion

3.1. Crystal structures

Fig. 1 shows the unit cells of $L1_0$-ordered MnGa, $D0_{22}$-ordered Mn_3Ga, and $L1_0$-ordered $Mn_{1.66}Ga$ alloys structures adopted in this work to perform systematic electronic structure calculations for pristine and strained structures. The unit cells of $L1_0$-ordered MnGa and $L1_0$-ordered $Mn_{1.66}Ga$, in Fig. 1(a) and (c), are doubled along the c axis for accurate quantitative comparison of results with the ones of $D0_{22}$-ordered Mn_3Ga unit cell. In $D0_{22}$-ordered Mn_3Ga unit cell, Fig. 1(b), manganese atoms that are located on the base square faces (1/2,1/2,0) and on the center plane (0,0,1/2) are denoted by Mn_I, and manganese atoms, with multiplicity 2, on the rectangular faces (0,1/2,1/4) are denoted by Mn_{II}. Ga atoms are located at the corners and center of the cuboid. The Mn_I atoms have a tetrahedral nearest neighbor environment. The magnetic moment of Mn_I is antiparallel to that of Mn_{II} in the ground state case. The Mn_I atoms in the $D0_{22}$ structure are replaced by Ga atoms in the $L1_0$-ordered MnGa unit cell, as shown in Fig. 1(a). One Ga atom in the $L1_0$-ordered MnGa unit cell is replaced by Mn_I atom in the $L1_0$-ordered $Mn_{1.66}Ga$ unit cell, as shown in Fig. 1(c).

The above crystal structures of MnGa alloys were optimization using the total energy minimization. The equilibrium lattice parameter has been computed by variation of the volume as well as the c/a ratio in the supercells. The optimized equilibrium lattice parameters of Mn–Ga alloys are given in Table 1. The equilibrium structural parameters are in very good agreement with experimental values that shown in Table 1 [20,28,45]. $L1_0$-MnGa lattice parameters, *a* and c, have small deviation of -0.2% from the experimental values. a lattice constant of $D0_{22}$-Mn_3Ga has deviations around -3.7%, while c lattice constant has deviations around $+0.87\%$ from the experimental values of the same structure. We compare the calculated lattice parameters of $L1_0$-$Mn_{1.66}Ga$ with the experimental lattice parameters of $L1_0$-$Mn_{1.6}Ga$ alloy [45], a and c lattice parameters have deviation of -3.1% and -0.39% from the experimental values of $L1_0$-$Mn_{1.6}Ga$ alloy. The deviations on the calculated lattice constants are typical for the DFT calculations.

3.2. Magnetocrystalline anisotropy

The magnetocrystalline anisotropy energy was calculated for Mn–Ga alloys in our study using force theorem. The MAE values are estimated at equilibrium structural parameters for the unstrained Mn–Ga alloys. The calculated value of MAE is equal 24.4 Merg/cm^3 for $L1_0$-MnGa, which is consistent with the theoretical value of 26 Merg/cm^3 and larger than the reported value around 19 Merg/cm^3 for the same structure [20,23]. The estimated value of MAE for $L1_0$- $Mn_{1.66}Ga$ is 22.6 Merg/cm^3 which is in very good agreement with the reported experimental value of 21.7 Merg/cm^3 in $L1_0$- $Mn_{1.5}Ga$ films [29], while larger than the reported value of 15 erg/cm^3 in $L1_0$-$Mn_{1.54}Ga$ films on MgO [13,46]. The calculated MAE for ground state structure of $D0_{22}$-Mn_3Ga is 27.3 Merg/cm^3, which is in good agreement with the previously estimated value using the linear muffin-tin orbitals method [13].

To study the effect of strain in MAE of Mn–Ga alloys, we vary the lattice parameters of the super cells of Mn–Ga structures and mapping the MAE values for different *a* and *c* lattice constants around the equilibrium values. Fig. 2 shows mapping of the MAE values of Mn–Ga alloys with respect to the different lattice parameters to represent the strain effect. As we can see in Fig. 2, MAE increases upon decrease in volume in MnGa alloy, while it is decreases in Mn_3Ga alloy. MAE in stoichiometric MnGa or Mn_3Ga compounds remains large when moderate strains are applied. In special case of bi-axial strain both compressive and tensile does not lead to significant change in MAE, the green lines in Fig. 2 represent the bi-axial strain (drawn to guide the eye. Assuming Poisson ratio ν is 0.3 $\Delta c/c = -2\nu/(1-\nu)^*\Delta a/a = -0.85^*\Delta a/a$). While tensile strain increases MAE in $Mn_{1.66}Ga$ alloys.