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First-principles study of electronic and magnetic properties and tetragonal distortion of the Heusler alloy Mn_2NiAl

Li-Jin Luo[a,*], Chong-Gui Zhong[a], Jing-Huai Fang[a], Peng-Xia Zhou[a], Yong-Lin Zhao[a], and Xue-Fan Jiang[b]

[a] *School of Science, Nantong University, Nantong 226007, China*
[b] *Jiangsu Key Laboratory of Advanced Functional Materials, Changshu Institute of Technology, Changshu 215500, China*

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Abstract. The crystal structure, tetragonal distortion, magnetism, electronic structure and pressure response of Mn_2NiAl are calculated by first-principles method based on the density functional theory. The calculations show, the equilibrium structure of Mn_2NiAl in the cubic austenitic phase is the MnMnNiAl structure with Mn atoms occupy A and B sites and two Mn atoms occupy inequivalent positions. In the process of transform from a cubic to a tetragonal structure, Mn_2NiAl alloys exhibit a stable martensitic phase near $c/a = 1.24$. In both the austenite and martensite phases, Mn atoms are the main contributors to the magnetism in Mn_2NiAl, Mn_2NiAl alloys show ferrimagnetism due to antiparallel but unbalanced magnetic moments of Mn(A) atom and Mn(B) atom. The direct d-d exchange interactions between Mn(A) atom and Mn(B) atom are weak because of small overlap of d-projected DOS of Mn(A) atom and Mn(B) atom nearby the Fermi level, but the intra-atomic interactions in Mn atoms are strong, this is the reason why the Mn_2NiAl alloys show ferrimagnetism. The findings strongly suggests that Mn_2NiAl alloys would behave like a magnetic shape memory alloy.

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Key words: first-principles, tetragonal distortion, martensite phase transformation, ferrimagnetism

1 Introduction

Magnetic shape memory alloy (MSMA) is a newly-developed shape memory material. It has not only thermoelastic shape memory effect as traditional shape memory alloy does by temperature control but also magnetic shape memory effect (MSME) by magnetic field control.

*Corresponding author. *Email address:* luolijin1964@126.com (L. J. Luo)

Compared with other intelligent driving materials like traditional shape memory alloy, piezoceramics, magnetostriction material, etc., MSMA has greater reinstate strain, output of stress and higher response frequency, making up the slowness of traditional shape memory alloy in response frequency and the shortage of piezoceramics and magnetostriction material in reinstate strain, and it is considered an ideal driving and sensing material, with a prospect of wide application.

Of the researches on MSMA, Heusler alloy is one of the materials which are conducted the most [1–3]. The findings have shown that a certain amount of Heusler alloys have MSME. The earliest finding, with regard to Heusler alloy with MSME that draws attention and stimulates deep research, is Ni_2MnGa [4–6]. Afterwards, Ni-Fe-Ga [7, 8], Co-Ni-Ga(Al) [9, 10], Ni-Mn-In(Sn, Sb) [11], etc., have been successively found with MSME.

Recent researches in theory and experiments have indicated that Mn_2NiGa, a new Heusler alloy, also has MSME [12], which exhibits a martensitic transformation around room temperature and an excellent two-way shape memory behavior with a strain of 1.7% in single crystal samples. It was also found that Mn_2NiGa has a Curie temperature up 588 K being much higher than that of studied Ni_2MnGa, whose Curie temperature is about 370 K. However, its ordered structure is different from the ordered structure of tradition $L2_1$ structure, i.e. Mn atoms occupy A and B sites, Ni atom C site and Ga atom D site, with a space group of $F\overline{4}3m$.

In this work, we mainly research another Heusler alloy Mn_2NiAl. Because it, like Mn_2NiGa, belongs to MnNi Heusler alloy, and Al and Ga belong to *sp* element as well, Mn_2NiAl may be considered a potential candidate for magnetic shape memory material. We investigate the crystal structure, tetragonal distortion, magnetism, electronic structure and pressure response of the Mn_2NiAl by first-principles method based on the density functional theory. The results show that Mn_2NiAl alloys exhibit a stable martensitic phase near $c/a = 1.24$ in the process of transform from a cubic to a tetragonal structure, and Mn_2NiAl alloys show ferrimagnetism in both austenite and martensite phases. These discoveries found strong evidence that Mn_2NiAl alloys would behave like a magnetic shape memory alloy.

2 Calculation method

The calculations are performed with the projected augmented wave (PAW) method based on the density functional theory, where the exchange-correlation potential is treated with the generalized gradient approximation of Perdew and Wang (PW91-GGA), the projected augmented wave (PAW) method is used in the interaction between ion and electron. The wavefunctions are expressed as a supposition of plane waves with a cut-off energy 400 eV. We choose k-points according to the scheme proposed by Monkhorst and Pack. A k-mesh parameters $19\times19\times19$ is adopted in the calculations of relaxation and static state, and $23\times23\times23$ is used in the calculations of DOS. The convergence criterion for total energy is 10^{-4} eV/au^3. The specific calculations are carried out with a package called VASP(Vienna abinitio simulation package) [13].

In the calculation, the structure of Mn_2NiAl in the cubic austenitic phase is taken to be