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Structure and magnetostriction of $Ho_{1-x}Pr_xFe_{1.9}$ alloys

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ABSTRACT

Polycrystalline $Ho_{1-x}Pr_xFe_{1.9}$ $(0 \leq x \leq 1)$ cubic Laves alloys were synthesized by arc-melting and subsequent annealing under high stress. Their structure, magnetic properties and magnetostriction are investigated using X-ray diffraction, vibrating sample magnetometer and standard strain gauge technique, respectively. It was found that $Ho_{1-x}Pr_xFe_{1.9}$ single cubic Laves phase cannot be obtained when $x > 0.2$ by a traditional vacuum annealing method. In contrast, the cubic Laves phase can be stabilized over the whole studies range in the samples annealed under high stress. The saturation magnetization for $Ho_{1-x}Pr_xFe_{1.9}$ decreases with the increase of x and reaches a minimum at $x = 0.4$, then increases with further increase of x, which indicates the antiparallel magnetic moment alignment between Ho and Pr sublattice. The magnetostriction of $Ho_{1-x}Pr_xFe_{1.9}$ does not linearly increase with increasing x, but presents a minimum at $x = 0.4$.

1. Introduction

RFe_2 (R = rare earths) C15 cubic Laves phase alloys were extensively investigated due to their giant magnetostriciton at room temperature. Terfenol-D ($Tb_{0.27}Dy_{0.73}Fe_2$), with low magnetic anisotropy and large low-field magnetostriction, has been widely applied in sonar transducer, sensors, actuators, etc. [1,2]. However, the raw materials of Terfenol-D are mainly expensive heavy rare earths Tb and Dy. According to the single-ion model, $PrFe_2$ possesses a larger calculated magnetostriction constant than $TbFe_2$ and $DyFe_2$ at 0 K due to its large second-order Stevens' factor α_J, ground state angular momentum J and average radius $\langle r_{4f}^2 \rangle$ of the 4f electron shell of the Pr^{3+} ion [1]. In addition, a magnetostrictive material with high-Pr content should have a good practical prospect because it is much cheaper than the heavy rare earths Tb or Dy. However, former studies showed that $PrFe_2$ cubic Laves phase could not be synthesized by conventional annealing at ambient pressure [3]. The unwanted non-cubic phase appears and the single phase materials cannot be synthesized in $(R,Pr)Fe_2$ alloys when Pr is over a certain concentration in rare earth sublattice [4–7]. Therefore, much attention has been paid to increase the concentration of Pr in $(R,Pr)Fe_2$ cubic Laves alloys. For example, Ren et al. recently reported the structure, magnetic and magnetostrictive properties in $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ compounds. They found that the introduction of a small amount of boron is beneficial to stabilize high-Pr content cubic Laves phase. But the single cubic Laves could not be obtained in $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ alloys when the Pr concentration exceeds 40 at% in rare-earth sublattice [6]. A similar result was also reported by Hari Babu et al. [7]. Up to present, $Ho_{1-x}Pr_xFe_2$ cubic Laves alloys have not yet been synthesized and their magnetic properties remain unknown. Recently, we reported that the structure and magnetic properties of $PrFe_x(1.5 \leq x \leq 3)$ alloys synthesized by annealing as-cast ingots under 6 GPa and the single cubic Laves phase was realized in $PrFe_{1.9}$ [8]. In this paper, polycrystalline alloys $Ho_{1-x}Pr_xFe_{1.9}$ $(0 \leq x \leq 1)$ with cubic Laves phase have been successfully synthesized by the same method. The crystal structure, magnetic properties and the magnetostriction of the alloys are investigated.

2. Experiment

Ingots with $Ho_{1-x}Pr_xFe_{1.9}$ ($x = 0.0$, 0.2, 0.4, 0.6, 0.8 and 1.0) stoichiometry were prepared by melting the appropriate constituent metals in a magneto-controlled arc furnace under a high-purity argon atmosphere. The as-cast ingots were pressed to 6 GPa by a hexahedral anvil press and heated to 900 °C for 30 min. Conventional X-ray diffraction (XRD) analysis was carried out using Cu Kα radiation with a Rigaku D/Max-gA diffractometer. The lattice parameters were calculated using the Jade 5.0 XRD analytical software (Materials Data, Inc., Livemore, CA). The magnetization measurements were carried out using a vibration sample magnetometer (7300, Lakeshore) under a magnetic field up to 10 kOe. The shape of sample for magnetostriction measurement was mainly disk-like with a diameter of 10 mm and a height

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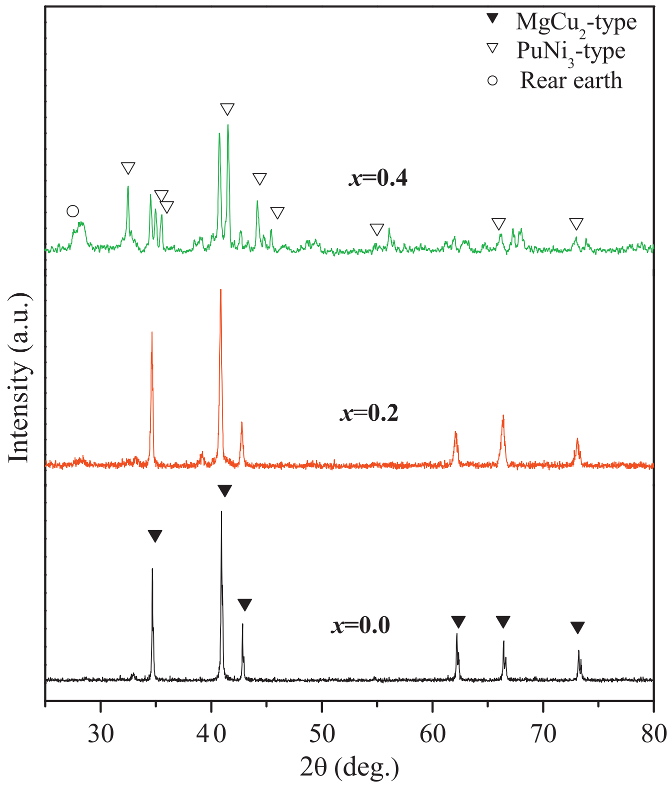

Fig. 1. XRD patterns of $Ho_{1-x}Pr_xFe_{1.9}$ alloys prepared by vacuum annealing at 900 °C for 30 min.

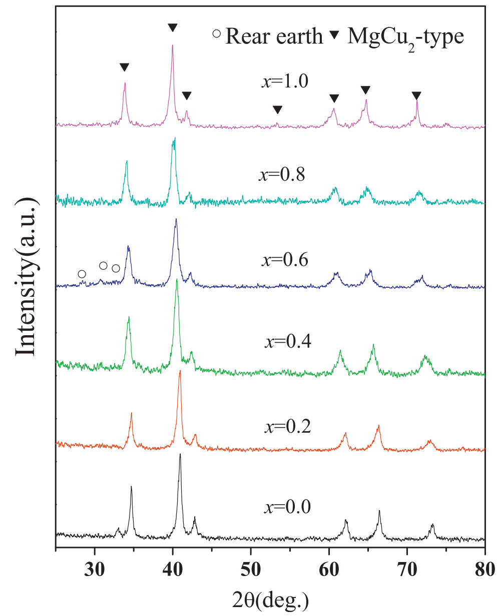

Fig. 2. XRD patterns of $Ho_{1-x}Pr_xFe_{1.9}$ alloys prepared by annealing under 6 GPa at 900 °C for 30 min.

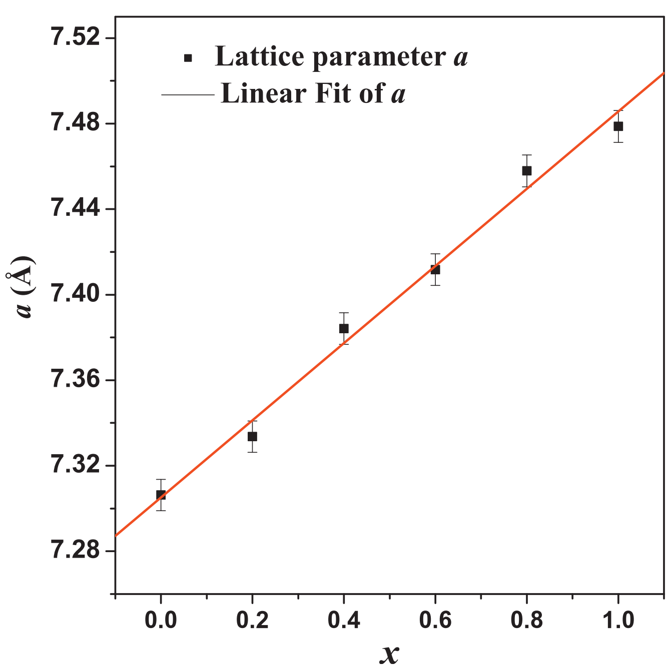

Fig. 3. The lattice parameter a of $Ho_{1-x}Pr_xFe_{1.9}$ Laves phase synthesized by annealing under 6 GPa at 900 °C for 30 min.

of 2 mm. The linear magnetostriction was measured using standard strain-gauge technique in directions parallel ($\lambda_{\parallel}$) and perpendicular ($\lambda_{\perp}$) to applied magnetic fields at room temperature.

3. Results and discussion

In order to study the structure of $Ho_{1-x}Pr_xFe_{1.9}$ alloys prepared by traditional high-temperature annealing method, the as-cast samples were vacuum annealed at 900 °C for 30 min. The XRD patterns for the samples with $x=0.0$, 0.2 and 0.4 are shown in Fig. 1. It can be observed that the single cubic Laves phase with $MgCu_2$-type structure can only be stabilized with $x \leq 0.2$. A multiphase structure appears when the Pr concentration further increases. For the sample with $x=0.4$, (Ho,Pr)Fe_3 with $PuNi_3$-type structure becomes the main phase and (Ho,Pr)Fe_2 with $MgCu_2$-type is the secondary phase, coexisting with a small amount rare earth phases. This is consistent with the structure of $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ alloys, in which the single cubic Laves phase cannot be stabilized when the Pr concentration is higher than 40 at% in rare-earth sublattice even with the help of Tb and B [6,7].

In contrast with Fig. 1, the samples prepared by annealing under high stress (Fig. 2) exhibit almost single cubic Laves phase with $MgCu_2$-type structure, coexisting with minor impurities, i.e. rare earth phases. Generally, the atomic size plays an important role in the formation of RFe_2 cubic Laves alloys. It was estimated that the ideal radius ratio between R and Fe for a cubic Laves phase is 1.225 [9]. As a result, it is difficult to obtain high-Pr content cubic Laves phases under normal pressure due to the large radius ratio between Pr and Fe. By the method of annealing at ambient pressure, the element of Ho with its smaller radius can stabilize $Ho_{1-x}Pr_xFe_{1.9}$ cubic Laves phase with Pr up to 20 at% in rare-earth sublattice, but cannot hold when the Pr concentration goes much higher. Therefore, the obtained stabilization of cubic Laves phase with high-Pr content in $Ho_{1-x}Pr_xFe_{1.9}$ should be attributed to the effects of high stress.

The lattice parameter (a) of $Ho_{1-x}Pr_xFe_{1.9}$ Laves phase synthesized under high stress is shown in Fig. 3. An approximate linear increase with Pr-concentration from 0.0 to 1.0 is found, as expected from Vegard's law: $a=xa_1+(1-x)a_2$, where a_1 and a_2 are the lattice parameters of $PrFe_{1.9}$ and $HoFe_{1.9}$, respectively. The increase of lattice parameter with increasing Pr concentration is due to the larger ionic radius Pr^{3+} than that of Ho^{3+}.

Magnetic field dependence of magnetization at 300 K for the $Ho_{1-x}Pr_xFe_{1.9}$ is shown in Fig. 4(a). We can note that each sample shows a tendency to saturation at the field of 10 kOe. For clarity, the magnetization at the maximum available 10 kOe, denoted as

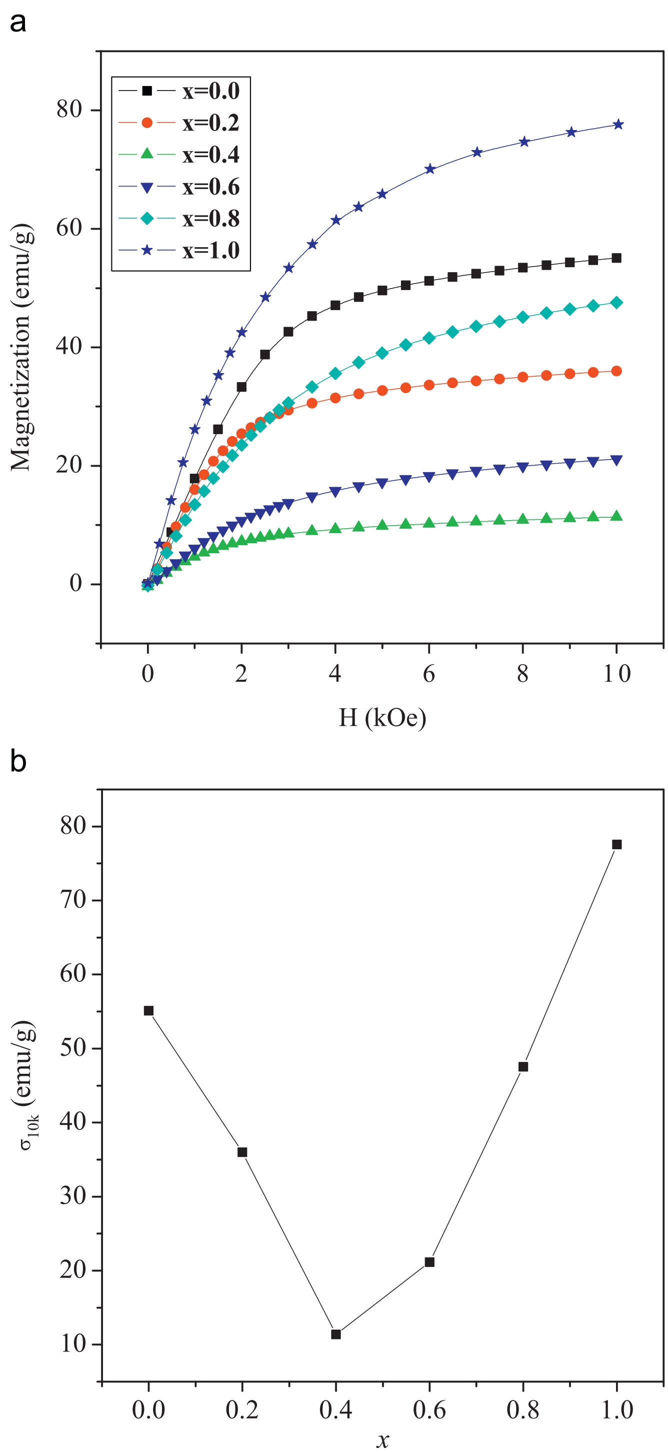

Fig. 4. (a) Magnetization curves of $Ho_{1-x}Pr_xFe_{1.9}$ cubic Laves alloys at 300 K and (b) the magnetization σ_{10k} at 10 kOe of $Ho_{1-x}Pr_xFe_{1.9}$ cubic Laves alloys at 300 K.

σ_{10k}, as a function of Pr concentration is plotted in Fig. 4(b). It can be seen that σ_{10k} decreases with increasing Pr concentration to a minimum at $x=0.4$, which is consistent with the variation of saturation magnetization for $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ alloys [6,7]. With further increasing x from 0.4 to 1.0, σ_{10k} increases monotonously. This can be understood by the compensation of sublattice magnetization: the moment of heavy rare earth Ho is unparallel that of Fe, whereas the moment of light rare earth Pr is parallel to. The magnetic moment of $Ho_{1-x}Pr_xFe_{1.9}$ can be described as: $\mu_s=(1-x)\mu_{Ho}-1.9\mu_{Fe}-x\mu_{Pr}$. As a result, the competition between these two sublattice moments leads to first decrease and following increase in the total net magnetization. This behavior could also be found in $Tb_{1-x}Pr_xFe_{1.9}$ and $Tb_{1-x}Nd_xFe_{1.9}$ alloy systems [10,11]. From the analysis the magnetization of the present alloy system, we can also get a conclusion that the magnetic moment compensation point between Ho and Pr should be in the range of $0.4\le x\le 0.6$ at room temperature.

The linear magnetostriction of all the samples was measured using standard strain-gauge technique in directions parallel ($\lambda_{\parallel}$)

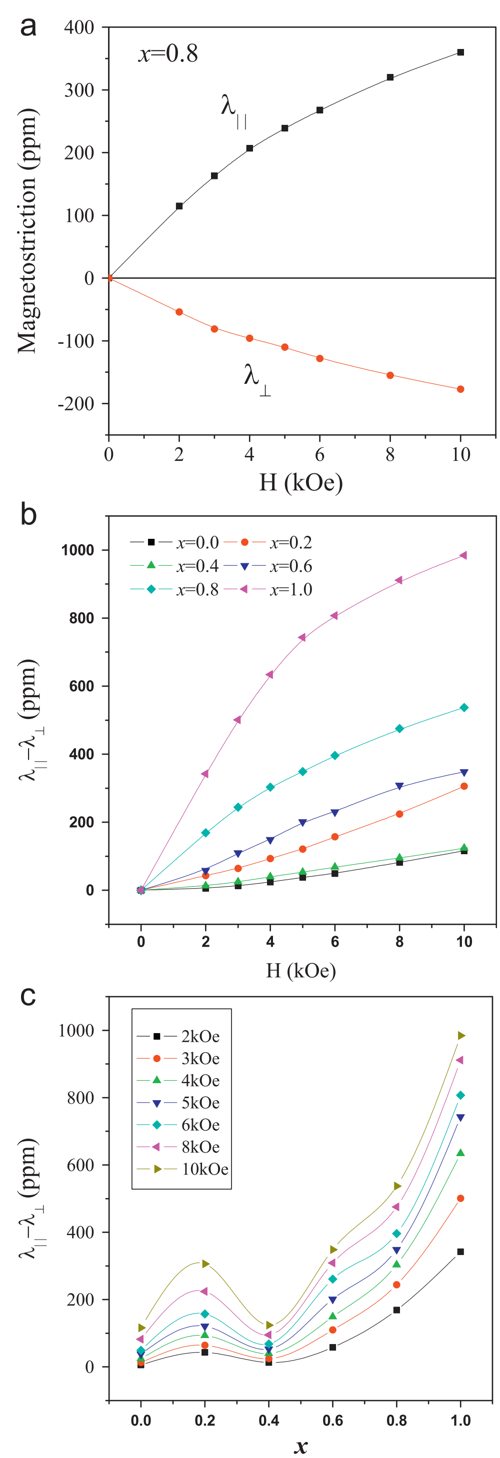

Fig. 5. (a) The magnetostrictions $\lambda_{\parallel}$ and $\lambda_{\perp}$ of $Ho_{0.2}Pr_{0.8}Fe_{1.9}$ versus the applied field, (b) the magnetostriction ($\lambda_{\parallel}-\lambda_{\perp}$) of $Ho_{1-x}Pr_xFe_{1.9}$ cubic Laves alloys versus the applied field and (c) the magnetostriction ($\lambda_{\parallel}-\lambda_{\perp}$) as a function of Pr concentration.

and perpendicular ($\lambda_\perp$) to applied magnetic fields at room temperature. As a typical representative, the magnetostrictions $\lambda_\parallel$ and $\lambda_\perp$ of $Ho_{0.2}Pr_{0.8}Fe_{1.9}$ versus the applied field are presented in Fig. 5(a). Fig. 5(b) shows the magnetostriction ($\lambda_\parallel - \lambda_\perp$) of $Ho_{1-x}Pr_xFe_{1.9}$ ($0 \le x \le 1$) versus the applied field. It is obvious that $HoFe_{1.9}$ possesses a much lower magnetostriction than $PrFe_{1.9}$. Besides, we can see that the magnetostriction of the sample with high-Ho content ($x \le 0.4$) is much harder to get saturation than that of high-Pr content. This indicates that $HoFe_{1.9}$ should have a larger anisotropy than $PrFe_{1.9}$.

In order to investigate the variation of magnetostriction with different Pr concentrations, the magnetostriction ($\lambda_\parallel - \lambda_\perp$) as a function of x is plotted in Fig. 5(c). The magnetostriction increases with the addition of Pr and shows a peak around $x = 0.2$. This tendency is very similar to that of $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ alloy system, in which the sample with $x = 0.2$ [6] or $x = 0.25$ [7] exhibits the largest magnetostriction. This is due to the sample with $x = 0.2$ or $x = 0.25$ is very close to the anisotropy compensation point between Ho^{3+} and Pr^{3+}. Consequently, the peak around $x = 0.2$ in the present studied alloys could be understood. With further increasing x to 0.4, the magnetostriction exhibits a minimum value in the whole range of magnetic fields, which is quite different from the magnetostriction variations of the well-known pseudobinary $Tb_xDy_{1-x}Fe_2$ or $Tb_xHo_{1-x}Fe_2$ system with two heavy rare-earths. This could be understood on the basis of the single-ion model: the saturation magnetostriction is proportional to M_{SR}, where M_{SR} is the saturation magnetization of the rare-earth sublattice [12]. Due to the antiparallel alignment of the Ho and Pr magnetic moments, $Ho_{0.6}Pr_{0.4}Fe_{1.9}$ has the lowest M_{SR} among the present studied alloy system. Therefore, the minimum magnetostriction for $Ho_{0.6}Pr_{0.4}Fe_{1.9}$ could be attributed to its lowest M_{SR}. A similar behavior could also be observed in some other heavy rare-earth and light rare-earth mixed pseudobinary alloys, such as $Tb_{0.4}Pr_{0.6}Fe_{1.9}$ [10] or $Tb_{0.4}Nd_{0.6}Fe_{1.9}$ [11]. With x increasing from 0.4 to 1.0, the magnetostriction of the system increases monotonically, which could be ascribed to the larger magnetostriction of $PrFe_{1.9}$ than that of $HoFe_{1.9}$.

4. Conclusion

High-Pr content $Ho_{1-x}Pr_xFe_{1.9}$ cubic Laves alloys which cannot be synthesized at ambient pressure were successfully fabricated by annealing under high stress. The structure, magnetic properties, and the magnetostriction of the alloys have been investigated. The variation of saturation magnetization for $Ho_{1-x}Pr_xFe_{1.9}$ is ascribed to the antiparallel moments between Ho and Pr. $PrFe_{1.9}$ possesses a much larger magnetostriction than $HoFe_{1.9}$. The magnetostriction does not linearly increase with increasing x, but presents a minimum at $x = 0.4$, which could be understood on the basis of single-ion model.

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References

[1] A.E. Clark, in: E.P. Wohlfarth (Ed.), Ferromagnetic Materials, vol. 1, North-Holland, Amsterdam, 1980, p. 531.
[2] N. Koon, C. Williams, B. Das, Journal of Magnetism and Magnetic Materials 100 (1991) 173.
[3] J.F. Cannon, D.L. Robertson, H.T. Hall, Materials Research Bulletin 7 (1972) 5.
[4] B. Wang, L. Cheng, K. He, Journal of Applied Physics 80 (1996) 6903.
[5] Z.J. Guo, S.C. Busbridge, B.W. Wang, Z.D. Zhang, X.G. Zhao, Journal of Applied Physics 85 (1999) 6677.
[6] W.J. Ren, D. Li, W. Liu, J. Li, Z.D. Zhan, Journal of Applied Physics 103 (2008) 07B311.
[7] V. Hari Babu, G. Markandeyulu, A. Subrahmanyam, Journal of Applied Physics 103 (2008) 07B313.
[8] Y.G. Shi, S.L. Tang, Y.J. Huang, B. Nie, B. Qian, L.Y. Lv, Y.W. Du, Journal of Alloys and Compounds 443 (2007) 11.
[9] D. Thoma, J. Perepezko, Journal of Alloys and Compounds 224 (1995) 330.
[10] Y.G. Shi, S.L. Tang, R.L. Wang, H.L. Su, Z.D. Han, L.Y. Lv, Y.W. Du, Applied Physics Letters 89 (2006) 202503.
[11] Y.G. Shi, S.L. Tang, Y.J. Huang, L.Y. Lv, Y.W. Du, Applied Physics Letters 90 (2007) 142515.
[12] E. Callen, H.B. Callen, Physical Review 139 (1965) A455.