Chinese Physics B

中国物理学会
Chinese Physical Society

ACCEPTED MANUSCRIPT

# Enhanced resonance frequency in $Co_2FeAl$ thin film with different thicknesses grown on flexible graphene substrate

To cite this article before publication: Cai Zhou *et al* 2023 *Chinese Phys. B* in press https://doi.org/10.1088/1674-1056/acfafa

Manuscript version: Accepted Manuscript

Accepted Manuscript is “the version of the article accepted for publication including all changes made as a result of the peer review process, and which may also include the addition to the article by IOP Publishing of a header, an article ID, a cover sheet and/or an ‘Accepted Manuscript’ watermark, but excluding any other editing, typesetting or other changes made by IOP Publishing and/or its licensors”

This Accepted Manuscript is **© 2023 Chinese Physical Society and IOP Publishing Ltd**.

During the embargo period (the 12 month period from the publication of the Version of Record of this article), the Accepted Manuscript is fully protected by copyright and cannot be reused or reposted elsewhere.

As the Version of Record of this article is going to be / has been published on a subscription basis, this Accepted Manuscript will be available for reuse under a CC BY-NC-ND 3.0 licence after the 12 month embargo period.

After the embargo period, everyone is permitted to use copy and redistribute this article for non-commercial purposes only, provided that they adhere to all the terms of the licence https://creativecommons.org/licences/by-nc-nd/3.0

Although reasonable endeavours have been taken to obtain all necessary permissions from third parties to include their copyrighted content within this article, their full citation and copyright line may not be present in this Accepted Manuscript version. Before using any content from this article, please refer to the Version of Record on IOPscience once published for full citation and copyright details, as permissions may be required. All third party content is fully copyright protected, unless specifically stated otherwise in the figure caption in the Version of Record.

View the article online for updates and enhancements.

This content was downloaded from IP address 113.57.237.69 on 26/12/2023 at 14:45

# Enhanced resonance frequency in $Co_2FeAl$ thin film with different thicknesses grown on flexible graphene substrate[*]

Cai Zhou(周偲),[1,3] Shaokang Yuan(袁少康),[2,3] Dengyu Zhu(朱登玉),[2,3] Yuming Bai(白宇明),[4] Tao Wang(王韬),[4] Fufu Liu(刘福福),[5] Lulu Pan(潘禄禄),[6] Cunfang Feng(冯存芳),[1,3] Bohan Zhang(张博涵),[1,3,†] Daping He(何大平)[7] and Shengxiang Wang(汪胜祥)[1, 3,‡]

[1] Hubei Engineering and Technology Research Center for Functional Fiber Fabrication and Testing, Wuhan Textile University, Wuhan 430200, P. R. China
[2] School of Electronic and Electrical Engineering, Wuhan Textile University, Wuhan 430200, P. R. China
[3] School of Mathematical and Physical Sciences, Wuhan Textile University, Wuhan 430200, P. R. China
[4] School of Integrated Circuits, Huazhong University of Science and Technology, Wuhan 430074, P. R. China
[5] Key Laboratory for Magnetism and Magnetic Materials, Ministry of Education, Lanzhou University, Lanzhou 730000, P. R. China
[6] Beijing National Center for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, P. R. China
[7] Hubei Engineering Research Center of RF-Microwave Technology and Application, Wuhan University of Technology, Wuhan 430070, P. R. China

The flexible materials exhibit more favorable properties than most rigid substrates in flexibility, weight saving, mechanical reliability, and excellent environmental toughness. Particularly, flexible graphene film with unique mechanical properties extensively explored in high frequency devices. Herein, we report the characteristics of structure and magnetic property at high frequency of $Co_2FeAl$ thin film with different thicknesses grown on flexible graphene substrate at room temperature. The exciting finding for columnar structure of $Co_2FeAl$ thin film laid the foundation of excellent high frequency property of $Co_2FeAl$/flexible graphene structure. In-plane magnetic anisotropy field varying with increasing thickness of Co2FeAl thin film can be obtained by measurement of ferromagnetic resonance, which can be ascribed to enhancement of crystallinity and increase of grain size. Meanwhile, the resonance frequency which can be achieved by measurement of vector network analyzer with the microstrip method increase with increasing thickness of Co2FeAl thin film. Moreover, in our case with graphene film, the resonance magnetic field is quite stable though folded it twenty cycles, which demonstrates that good flexibility of graphene film and the stability of high frequency magnetic property of $Co_2FeAl$ thin film grown on flexible graphene substrate. These results are promising for the design of microwave devices and wireless communication equipment.

**Keywords:** Enhanced resonance frequency, Magnetic resonance field, flexible graphene substrate
**PACS:** 32.30.Dx, 75.70.-I, 76.50.+g

---

[*] This work is supported by the National Natural Science Foundation of China (Grant Nos. 51901163 and 12104171), the Fundamental Research Funds for the Central Universities (Grant No. 2021XXJS025).

[†] Corresponding author. E-mail: bhzhang@wtu.edu.cn
[‡] Corresponding author. E-mail: shxwang@wtu.edu.cn

Accepted Manuscript

# 1. Introduction

Recently, with improvement in the standard of living, the flexible electronic devices have received wide attention owing to perfect stretchability, low cost, biocompatibility, and light weight, which have great potential in application of sensitive skin, flexible circuit boards, and paper-like displays. In these flexible materials, the advanced carbon material-based films, including carbon fibers,[1] carbon nanotubes,[2-4] multi-shelled fullerenes, [5,6] and especially graphene, [7,8] exhibit more favorable properties than most rigid substrates in flexibility, weight saving, mechanical reliability, and excellent environmental toughness. The flexible graphene films can be extensively explored in high frequency devices due to the unique mechanical properties. [9-11] He's group has reported flexible graphene films for radio-frequency antennas, multi-beam radiation, highly sensitive wearable sensor, etc. [12-14] In fact, the graphene film as flexible substrate is expected to Heusler metallic materials are considered to be ideal compounds as high spin polarized current sources, and some of them generally have a high Currie temperature, high spin polarization and low damping, [15,16] Among the Heusler metallic materials, with the formula of $X_2YZ$ (where X is a transition metal element, Y is another transition metal element, and Z is a main group sp element), particularly $Co_2FeAl$ (CFA), have attracted intense research interest due to the half-metals even at room temperature. [17-19] Belmeguenai's research group have investigated the static and dynamic magnetic property in CFA thin films by sputtering on a Si and MgO substrates annealed at different temperatures. [20] Our group have reported the electric field tuning magnetic anisotropy in CFA thin film grown on PMN-PT substrate. [21,22] In these cases, the CFA thin film were fabricated on the above conventional rigid substrate. This greatly limits the application scope of flexible thin film materials. Therefore, it is essential to study the magnetic properties of magnetic thin films fabricated on flexible substrate. In this work, the high frequency property of CFA thin film with different thicknesses grown on flexible graphene substrate (FGS) is analyzed in detail at room temperature. With the thickness of CFA thin film grown on FGS varying from 20 nm to 200 nm, the increase of grain size can be obtained. The in-plane magnetic anisotropy field decreases with increasing the thickness of CFA thin film from 20 nm to 100 nm measured by ferromagnetic resonance, which can be attributed to the change of structure of CFA thin film with different thicknesses grown on FGS. However, in-plane magnetic anisotropy field increases with continuing to increase thickness to 200 nm, which can lead to the resonance frequency shifting to higher frequency measured by using a vector network analyzer with the microstrip method.

# 2. Experimental details

The CFA thin films with different thicknesses (20, 50, 100, 150, and 200 nm) were deposited by Direct Current (DC) magnetron sputtering on FGS at a common base pressure < $8\times10^{-4}$ Pa and work processing Ar pressure of 0.1 Pa, a power of 150 W. A field emission scanning electron microscope (SEM, Hitachi SU5000) was employed to observe the cross-section microstructures and the surface morphologies. The x-ray diffraction (XRD) measurements were performed on X'Pert x-ray powder diffractometer with $CuK_\alpha$ radiation (1.54056Å). The ferromagnetic resonance (FMR) measurements were carried out with a PNA 8722ES vector network analyzer using the coplanar waveguide method. Permeability spectra were carried out with a PNA E8363B vector network analyzer using the microstrip method.

# 3. Results and discussion

The cross-sectional and highly magnified SEM images of the FGS show that the film based on orderly

Accepted Manuscript

stacked graphene layers with a thickness of 108 μm as shown in Fig. 1(b) and Fig. 1(c). The cross-section image of CFA thin film with different thicknesses grown on FGS is also observed by SEM. For brevity, only the 200 nm-thick CFA grown on FGS is shown in Fig. 1(a). The columnar structure of CFA can be obviously observed, which is comparable to the sample grown on Si substrate. [23,24] The exciting finding laid the foundation of the good quality of CFA/FGS structure. In addition, surface morphologies of CFA/FGS structure with various thickness were shown in the inset of Fig. 2(b). Firstly, the smooth surface morphologies of FGS can be obtained as shown in the top inset of Fig. 2(b). Then, we can observe that with increasing thickness of CFA thin film, the grain continues growth and corresponding size becomes larger. This result indicates thickness has a great influence on microstructure for CFA thin film grown on FGS.

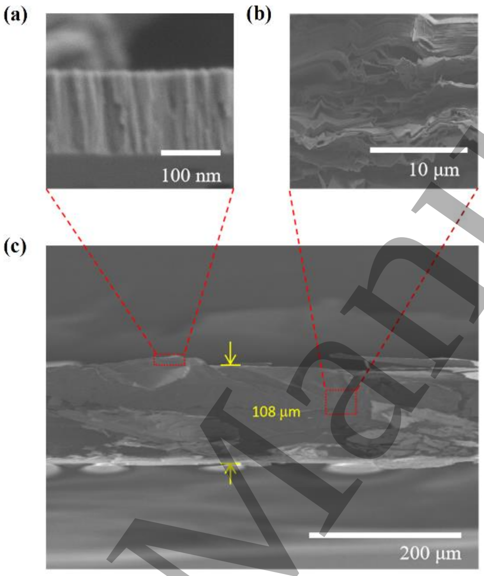

**Fig. 1.** The SEM cross-section images of (a) CFA thin film, (b) FGS and (c)CFA/FGS structure, respectively.

A piece of rolled FGS was shown in the inset of Fig. 2(a), which demonstrates FGS have good flexibility. The crystal structures for all thin films were measured by XRD as shown in Fig. 2(a).The sharp diffraction peak (002) and peak (004) of FGS is located at 26.3 ° and 54.5 °. The (022) peak of CFA thin film appears at 44.7 °. Moreover, the intensity of (022) peak of CFA thin film increases with increasing thickness of CFA thin film as shown in the top inset of Fig. 2(a), which indicates the enhancement of crystallization. However, the position of (022) peak of CFA thin film remains unchanged. According to the Scherrer Equation, grain sizes of CFA thin films were 28.6 nm, 29.7 nm, 31.1 nm, 31.8 nm and 34.2 nm with increasing thickness of CFA thin films, respectively, as shown in Fig. 2(b), which is consistent with variation tendency of surface morphologies measured by SEM as shown in the inset of Fig. 2(b). The increase of grain size with increasing thickness of CFA thin film can indirectly influence the change of in-plane magnetic anisotropy field. The above results confirmed the good quality of CFA/FGS structure we prepared, which have huge impact on the physical properties, especially high-frequency magnetic properties.

Accepted Manuscript

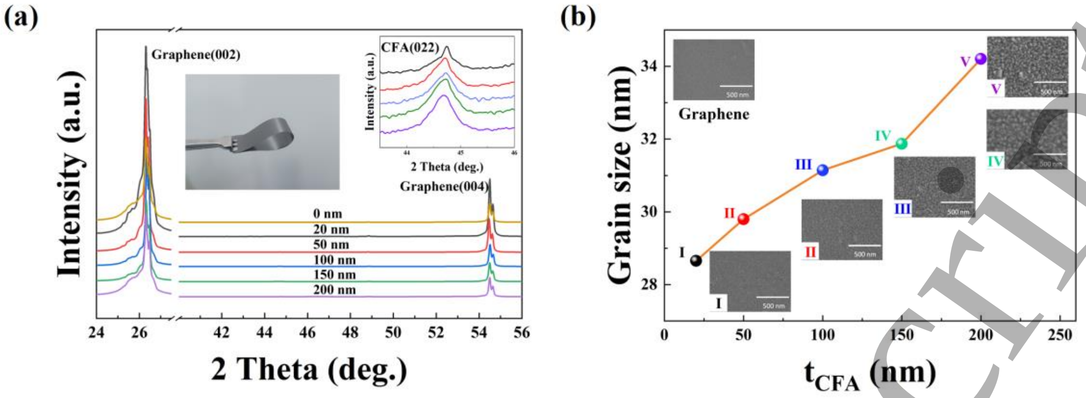

**Fig. 2.** (a) XRD patterns of CFA/FGS structure with different thicknesses of CFA thin film and (0 nm) pure graphene substrate. The inset shows digital photograph of FGS; (b) The grain size versus thickness of CFA thin film. The inset images show surface morphologies of CFA/FGS structure with different thicknesses and FGS.

The high-frequency magnetic properties of CFA thin films were measured by ferromagnetic resonance (FMR) using a vector network analyzer with coplanar waveguide method as shown in Fig. 3(a) and in the top inset of Fig. 3(c). In order to prove the good flexibility of graphene film and the stability of high frequency magnetic property of $Co_2FeAl$ thin film grown on flexible graphene substrate, we choose the sample with 150-nm thick CFA thin film to fold over twenty cycles and then take FMR measurement at fixed 10 GHz every five cycles. The folding schematic diagram is shown in the bottom inset of Fig. 3(b). As shown in Fig. 3(b), the curves basically remain unchanged within twenty cycles. The resonance magnetic field approximately 800 Oe is obtained by fitting these curves according to Eq. (1). However, the dramatic change of the FMR curve occurs in folding twenty-fifth cycles. The shape of curve becomes slightly shaking and the curve deforms, which indicates the quality of CFA thin film deteriorates after being folded twenty cycles. The more cracks can be distinctly observed after twenty cycles as shown in the top inset of Fig. 3(b), which can be measured by RX50M Series Metallurgical Microscope. The similar result can be obtained for CFA thin film with 20 nm, 50 nm, 100 nm and 200 nm. Fig. 3(c) shows under different frequencies the typical FMR spectra for 150 nm-thick CFA thin film. In general, the magnetization is probed using a special phase correlation under the microwave excitation, and the FMR spectrum does not correspond to the imaginary part of the susceptibility alone, but in fact represents a mixture of the imaginary and real parts. Therefore, the actual function of the absorption curve can be fitted by an asymmetric Lorentzian function [25]:

$$\varphi(H) = \mathrm{A}\frac{\Delta H\cos\delta + (H - H_r)\sin\delta}{\Delta H^2 + (H - H_r)^2} \qquad (1)$$

Where $H$ is the external magnetic field, $H_r$ is the magnetic resonance field, $\Delta H$ is the half-width at half-maximum of the linewidth, $\delta$ is the phase that mixes the real and imaginary parts of the dynamic susceptibility, and A is the integral coefficient. The fitting solid curve is shown in Fig. 3(c) in light gray color. The fitting result reveals that $H_r$ shifts towards higher magnetic field with increasing frequency. For 150 nm-thick CFA thin film, the resonance magnetic field increase from 0.258 kOe to 1.268 kOe with the microwave frequency increasing from 6 GHz to 12 GHz as shown in Fig. 3(c). In addition,

Accepted Manuscript

frequency-dependent $\varphi(H)$ curves were measured for all thin film. As a result, the data of frequency-dependent $H_r$ under different thicknesses of CFA thin film are shown in Fig. 3(d), which is fitted using the Kittel's equation. [26]

$$f = \frac{\gamma}{2\pi}\sqrt{(H_r + H_k)(H_r + H_k + 4\pi M_s)} \qquad (2)$$

Where gyromagnetic ratio $\gamma/2\pi$=2.8 GHz/kOe [27,28], $4\pi M_s$ is the saturation magnetization, and $H_k$ is the in-plane magnetic uniaxial anisotropy field. The data of frequency-dependent $H_r$ was fitted using Equation (2) to extract $4\pi M_s$ and $H_k$. The fitting curve was shown in Fig. 3(d) with red color. With increasing thickness of CFA thin film, the value of $4\pi M_s$ is almost unchanged. However, $H_k$ is found to decrease from 0.243 kOe to 0.062 kOe with increasing thickness from 20 nm to 100 nm as shown in Fig. 4(a). This can be explained as following: The crystallization of CFA thin film is enhanced with the increase of thickness of CFA thin film as shown in the inset of Fig. 2(b). Meanwhile, the grain size become larger. The competition between uniaxial magnetic anisotropy and magnetocrystalline anisotropy can appear, which can lead to the decrease of in-plane magnetic uniaxial anisotropy field. Nevertheless, $H_k$ increases from 0.134 kOe to 0.180 kOe with thicknesses increasing from 150 nm to 200 nm. The sudden increase in $H_k$ for CFA thin film with 150 nm-thick can be attributed to releasing of substrate induced strain[29], which can lead to the higher resonance frequency.

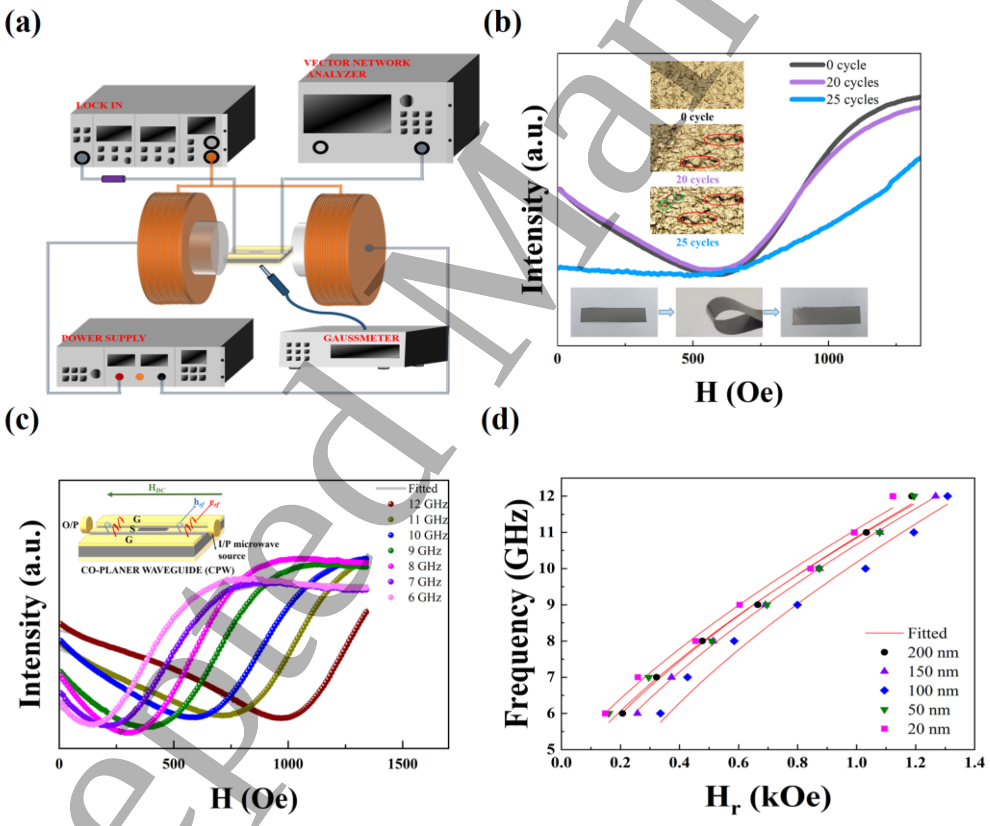

**Fig. 3.** (a) The schematic illustration of ferromagnetic resonance (FMR) using a vector network analyzer; (b) These curves under unfolded state, and after being fold twenty cycles and twenty-fifth cycle. The top inset shows the more cracks with increasing folding cycles. The bottom inset shows the folding schematic diagram; (c) The experimental data (dots) and fitting curves (lines) of FMR for 150 nm-thick CFA thin film. The inset shows the device schematic of microstrip method; (d) The applied microwave frequency versus resonance magnetic field with different thicknesses of CFA thin film.

Accepted Manuscript

The dependence of permeability on the frequency with different thicknesses of CFA thin film was measured using a vector network analyzer with the microstrip method, as shown in the bottom inset of Fig. 4(b). When the thickness of CFA thin film less than 100 nm, there is no resonance peak detected in this method, which can be attributed to the disorderly arrangement of magnetic moment. This is related to roughness of surface morphologies. With continuing to increase the thickness of CFA thin film, the resonance peak can be obviously observed for CFA thin film with 150 nm and 200 nm-thick. The resonance frequency enhances from 3.4 GHz to 5.6 GHz, which is consistent with the result of $H_k$ -dependent thickness of CFA thin film. Moreover, the resonance magnetic field and resonance frequency are quite stable though folded it over ten times, which demonstrates that good flexibility of graphene film and the ultrastability of high frequency property of CFA thin film grown on FGS.

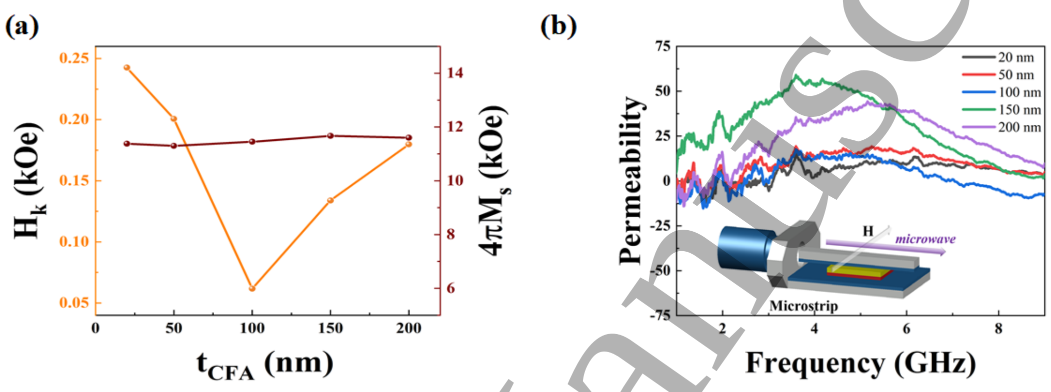

**Fig. 4.** (a) The change of $H_k$ and $4\pi M_s$ with different thicknesses of CFA thin film; (b) The permeability spectra of CFA thin film. The inset shows the device schematic of coplanar waveguide method

## 4. Conclusion

In summary, we identified characteristics of CFA/FGS structure and high frequency magnetic property of CFA thin film with different thicknesses grown on FGS. With increasing thickness of CFA thin film, the surface becomes smoother, and grain size gradually increase, which can influence the change of in-plane magnetic anisotropy field. The in-plane magnetic anisotropy field gradually decreases when the thickness of CFA thin film increases from 20 nm to 100 nm by measurement of FMR. However, with thickness increasing to 100 nm, in-plane magnetic anisotropy field suddenly increases. The measurement result of the dependence of permeability on the frequency demonstrates that the resonance peak can be obviously observed at 3.4 GHz and 5.6 GHz with thickness 150 nm and 200 nm, which is in agreement with the result of FMR. Moreover, the high frequency parameter remains stable though folded it over twenty cycles, which indicates that good flexibility of graphene film and the ultrastability of high frequency property of CFA thin film grown on FGS. These results can be used to remove the obstacles that communication system transmission distance and signal quality.

## References

[1] Mehdlpour A, Trueman C W, Sebak A R, and Hoa S V 2009 *IEEE Antennas and Propagation Society International Symposium*, June 1-5, 2009, North Charleston, SC, USA, p. 1

Accepted Manuscript

[2] Rutherglen C, Jain D and Burke P 2009 *Nat. Nanotechnol.* **4** 811

[3] Elwi T A, Al-Rizzo H M, Rucker D G, Dervishi E , Li Z and Biris A S 2010 *Nanotechnology* **21** 045301

[4] Puchades I, Rossi J E, Cress C D, Naglich E and Landi B J 2016 *ACS Appl. Mater. Interfaces.* **8** 20986

[5] Vacirca N A, McDonough J K, Jost K, Gogotsi Y and Kurzweg T P 2013 *Appl. Phys. Lett.* **103** 073301

[6] Shenderova O, Grishko V, Cunningham G, Moseenkov S, McGuire G and Kuznetsov V 2008 *Diam. Relat. Mater.* **17** 462

[7] Sajal S Z, Braaten B D and Marinov V R 2015 *IEEE International Symposium on Antennas and Propagation & USNC/URSI National Radio Science Meeting*, July 19-24, 2015, Vancouver, BC, Canada, p. 2415

[8] Sa'don S N H, Kamarudin M R, Ahmad F, Jusoh M and Majid H A 2017 *Appl. Phys. A* **123** 118

[9] Takeichi T, Eguchi Y, Kaburagi Y, Hishiyama Y and Inagaki M 1999 *Carbon* **37** 569

[10] Ubbelohde A R 1976 *Carbon* **14** 1

[11] Hishiyama Y, Nakamura M, Nagata Y and Inagaki M 1994 *Carbon* **32** 645

[12] Fan C, Wu B, Song R G, Zhao Y T, Zhang Y H and He D P 2019 *Carbon* **155** 506

[13] Song R G, Mao B Y, Wang Z, Hui Y Y, Zhang N, Fang R, Zhang J W, Wu Y, Ge Q, Novoselov K S and He D P 2023 *PNAS* **120** e2209807120

[14] Qian W, Fu H Q, Sun Y, Wang Z, Wu H, Kou Z K, Li B W, He D P and Nan C W 2022 *Adv. Mater.* **34** 2206101

[15] de Groot R A, Mueller F M, van Engen P G and Buschow K H J 1983 *Phys. Rev. Lett.* **50** 2024

[16] Felser C and Hillebrands B 2007 *J. Phys. D: Appl. Phys.* **40** E01

[17] Gabor M S, Petrisor T, Tiusan C, Hehn M and Petrisor T 2011 *Phys. Rev. B* **84** 134413

[18] Cui Y S, Khodadadi B, Schäfer S, Mewes T, Lu J W and Wolf S A 2013 *Appl. Phys. Lett.* **102** 162403

[19] Gupta R, Husain S, Kumar A, Brucas R, Rydberg A and Svedlindh P 2021 *Adv. Opt. Mater.* **9** 2001987

[20] Belmeguenai M, Tuzcuoglu H, Gabor M S, Petrisor Jr T, , Tiusan C, Zighem F, Chérif S M and Moch P 2014 *J. Appl. Phys.* **115** 043918

[21] Zhou C, Dunzhu G S, Yao J L and Jiang C J 2017 *J. Alloy Compd.* **710** 680

[22] Zhou C, Wang F L, Dunzhu G S, Yao J L and Jiang C J 2016 *J. Phys. D: Appl. Phys.* **49** 455001

[23] Wang G X, Dong C H, Wang W X, Wang Z L, Chai G Z, Jiang C J and Xue D S 2012 *J. Appl. Phys.* **112** 093907

[24] Zhou C, Wang F L, Wei W W, Wang G X, Jiang C J and Xue D S 2013 *J. Phys. D: Appl. Phys.* **46** 425002

[25] Mecking N, Gui Y S and Hu C M 2007 *Phys. Rev. B* **76** 224430

[26] Kittel C 1948 *Phys. Rev. B* **73** 155

[27] Song N N, Gu S Z, Wu Q, Li C L, Zhou J, Zhang P P, Wang W and Yue M 2018 *J. Magn. Magn. Mater.* **451** 793

[28] Holanda J, Saglam H, Karakas V, Zang Z Z, Li Y, Divan R, Liu Y Z, Ozatay O, Novosad V, Pearson J E and Hoffmann A 2020 *Phys. Rev. Lett.* **124** 087204

[29] Kipgen L, Fulara H, Raju M and Chaudhary S 2012 J. Magn. Magn. Mater. **324** 3118

Accepted Manuscript