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Cite as: AIP Advances 6, 025027 (2016); <https://doi.org/10.1063/1.4943082>

Submitted: 09 June 2015 . Accepted: 18 February 2016 . Published Online: 26 February 2016

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Optimum thickness of soft magnetic phase in FePt/FeCo permanent magnet superlattices with high energy product and large magnetic anisotropy energy

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(Received 9 June 2015; accepted 18 February 2016; published online 26 February 2016)

Ab initio calculations on hard/soft (FePt)_m/(FeCo)_n, (m = 4, 6, 8 and n = 2-2m) magnetic superlattices show that the B2 type FeCo layers become anisotropic with varying interlayer spacing and enhanced magnetic moments. The average magnetic moment in superlattices is higher than in bulk FePt, resulting in high maximum energy product for (FePt)₄/(FeCo)₈ which is nearly double the calculated value for bulk FePt. The calculation of the magnetic anisotropy energy shows that the optimal thickness of the soft magnetic phase for good permanent magnet behaviour of the superlattice is less than ~2 nm. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4943082>]

I. INTRODUCTION

Permanent magnets are of vital importance for a wide range of applications including household gadgets, data recording media, medical applications, motors, and power generation equipments.¹ The strongest known magnets have rare earths (REs) such as Nd and Sm that have helped to reduce the size of many electronic devices. There is great interest to develop RE free strong magnets with large coercivity and high magnetization using exchange coupled composites and superlattices. A key figure of merit is the maximum energy product $(BH)_{\max} = \mu_0 M_s^2 / 4$, where M_s is the saturation magnetization and μ_0 is the permeability. RE based magnets SmCo₅ and Nd₂Fe₁₄B have a high energy product^{2,3} of 31 MGOe and 50 MGOe, respectively but the value for RE free magnets is generally lower. Here we report from calculations a high value (up to ~97 MGOe) of $(BH)_{\max}$ for FePt/FeCo exchange coupled superlattices and show that the optimal thickness of the soft magnetic phase to achieve high magnetization as well as large magnetic anisotropy energy (MAE) is less than ~2 nm. It is noteworthy that similar conclusions have been drawn from recent experiments on FePt/FeCo thin films^{4,5} as well as FePt/Fe₃O₄ nanoparticle composites.⁶

Kneller and Hawig⁷ proposed exchange coupled permanent magnets using a combination of hard and soft magnets to achieve high magnetic anisotropy and high saturation magnetization. The hard magnet induces anisotropy in the soft magnet and this effect was suggested to be pronounced when the soft magnetic region is smaller than twice the domain wall width of the hard magnet which is typically 4-5 nm.⁸ Early model calculations⁹ suggested the width of the soft magnetic phase to be less than 9 nm while recent model calculations¹⁰ suggested the width of the hard magnetic phase to be ~2 nm. We use *ab initio* calculations to understand the atomic structure as well as magnetic anisotropy in such systems and focus on FePt/FeCo superlattices as thin films of FePt/Fe, FePt/Co, and FePt/FeCo have been realized^{4,5} and a high value of 54 MGOe for the energy product has been obtained¹¹ for aligned and exchange-coupled FePt based films. Also a similar value of 50 MGOe has been obtained⁵ for films with 5 monolayers (ML) of FeCo on 7 ML of FePt.

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Earlier Hong *et al.*^{12,13} have studied FeCo/FePt superlattices from *ab initio* calculations by considering L1₀-ordered FePt and fcc type FeCo layers. However, the crystal structure of FeCo is B2-type and recently Wang *et al.*⁴ have grown FeCo layers epitaxially on FePt and confirmed the B2 structure. Thus prepared Fe₆₀Pt₄₀/Fe₄₀Co₆₀ bilayers have higher anisotropy field and higher MAE in comparison to Fe₆₀Pt₄₀/Fe and Fe₆₀Pt₄₀/Co bilayers. The optimal thickness of the FeCo layers was found to be less than 2 nm to produce high anisotropy field. The easy axis of FeCo was found to be normal to the film plane when its thickness was < 3 nm. Interestingly, Fe/Co layers grown on AuCu (001) buffer also develop perpendicular anisotropy with energy density of about $1 \times 10^7 \text{ erg/cm}^3$ in a 3 nm thick sample.¹⁴ Here we study superlattices with L1₀-ordered FePt and B2-type FeCo layers by varying the thicknesses of the hard (FePt) and soft (FeCo) magnetic layers. Our results suggest large uniaxial anisotropy constant K_u as well as large magnetization when the thickness of the soft magnetic phase is less than ~ 2 nm. This result is similar to the conclusions drawn from experiments on thin films.^{4,5}

II. METHODOLOGY

We constructed m-n superlattices by arranging m layers of FePt and n layers of FeCo in such a way that the L1₀ ordered FePt layers are rotated by 45° with respect to B2-FeCo layers so that the nearest neighbour in-plane distance in FePt layers matches with the in-plane lattice parameters of the B2-type FeCo layers as the difference between the two values is small. We considered m = 4, 6, and 8 and varied n from 2 to 2m so that the stoichiometries of FePt and FeCo layers remained intact. In Fig. 1 we have shown a supercell for 4-8 superlattice. Similar models have been designed for other superlattices. We used projector augmented wave pseudopotential method in Vienna *Ab initio* Simulation Package¹⁵ and generalized gradient approximation¹⁶ for the exchange-correlation functional. The cut-off energy for the plane waves was taken to be 366.55 eV and $9 \times 9 \times k$ (k = 1 and 3) Monkhorst-pack¹⁷ k-points mesh was used to achieve structural optimization. All the ions as well as lattice parameters were relaxed without using any constraint until the absolute value of the force on each ion became smaller than 0.005 eV/\AA . The electronic minimization was performed with an accuracy of 10^{-5} eV. As a test, the optimized lattice parameters for bulk FePt were calculated to be $a = 3.855 \text{ \AA}$ and $c = 3.772 \text{ \AA}$, and for B2-FeCo, $a = 2.841 \text{ \AA}$ which are in very good agreement with the experimental values of

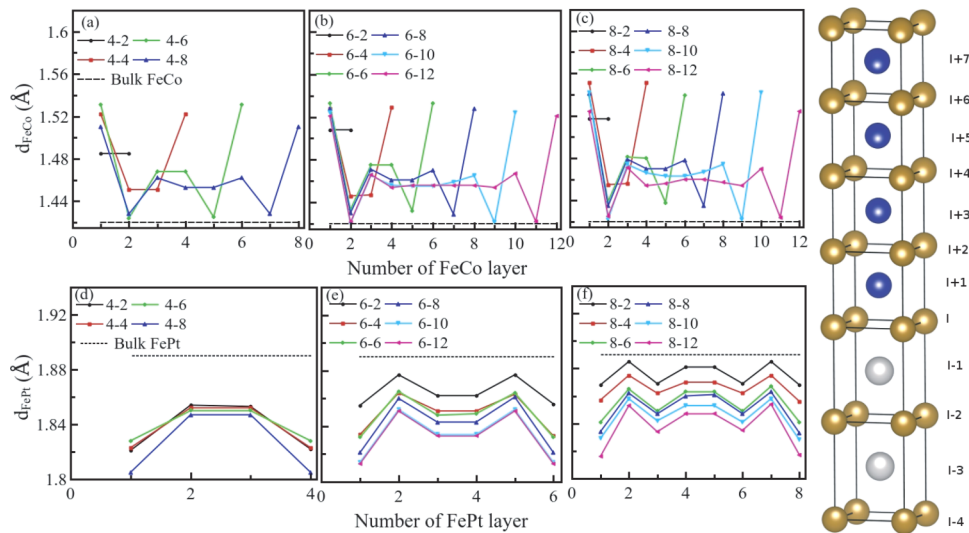


FIG. 1. Variation in the interlayer separations of FeCo and FePt layers starting with the first layer from the interface. (a) and (d) correspond to $m = 4$, (b) and (e) $m = 6$, and (c) and (f) $m = 8$ with different values of n . Dashed line indicates the interlayer separation of bulk FeCo (1.42 \AA) and bulk FePt (1.886 \AA). A ball and stick model of $(\text{FePt})_4/(\text{FeCo})_8$ superlattice with 4 layers of FePt in L1₀ structure and 8 layers of FeCo in B2 structure is also shown. Brown, grey, and blue balls represent Fe, Pt, and Co atoms, respectively. The FePt layers are rotated by 45° with respect to FeCo layers. The symbol I refers to the interface layer.

$a = 3.86 \text{ \AA}$ and $c = 3.788 \text{ \AA}$ for FePt and $a = 2.84 \text{ \AA}$ for FeCo. Calculations of MAE were carried out by including spin-orbit coupling for [100] and [001] directions of the spin orientation, [001] being the direction of the superlattice. For this we used 1280 (16x16x5) \mathbf{k} -points in the case of 4-2, 4-4, and 6-2 superlattices, 768 (16x16x3) \mathbf{k} -points for 4-6, 4-8, 6-4, 6-6, 6-8, 6-10, 6-12, 8-2, 8-4, 8-6, 8-8, and 8-10 superlattices, and 256 (16x16x1) \mathbf{k} -points for 8-12 to 8-16 superlattices in order to maintain approximately equal density of \mathbf{k} -points. The tolerance for the energy convergence is taken to be 10^{-7} eV. Our calculated MAE for bulk FePt using 1728 (12x12x12) \mathbf{k} points is 2.14 meV per formula unit (fu) and it is comparable to the values¹⁸⁻²⁴ (2.22 - 2.98 meV/fu) obtained in earlier calculations. However, it is larger compared with the experimental value²⁵ of 1.2 meV/fu. Further we calculated the uniaxial anisotropy constant K_u which is the difference in energies for the cases with spin orientation in [100] and [001] directions divided by the cell volume. The calculated K_u with the easy axis along [001] direction for bulk FePt is $12.2 \times 10^7 \text{ erg/cm}^3$. This is consistent with the earlier reported theoretical value^{26,27} but it is overestimated compared with the experimental value²⁸ of $6.6 \times 10^7 \text{ erg/cm}^3$. The difference from experimental values can also arise from the fact that our calculations are on an ideal system while in reality there would be defects as well as concentration variation that will also affect the values.

III. RESULTS AND DISCUSSION

In the optimized structures of the superlattices, the interlayer separations d_{FePt} and d_{FeCo} as well as the in-plane lattice parameters a and b ($a = b$) vary as shown in Figs. 1 and 2(a) for different superlattices. In general, for a given n , a is reduced towards the value of bulk FePt if the number of FePt layers is increased while for a given m , a increases towards the value for FeCo as n is increased. This is because the in-plane nearest neighbour bond length in FePt (2.726 \AA) is slightly shorter than the lattice constant of bulk FeCo (2.841 \AA). There is an oscillatory behavior of d_{FePt} and d_{FeCo} similar to that known on free surfaces with the largest contraction (expansion) for the first layer of FePt (FeCo) from the interface (see Fig. 1). For the next layer from the interface, d_{FePt} (d_{FeCo}) increases (decreases)

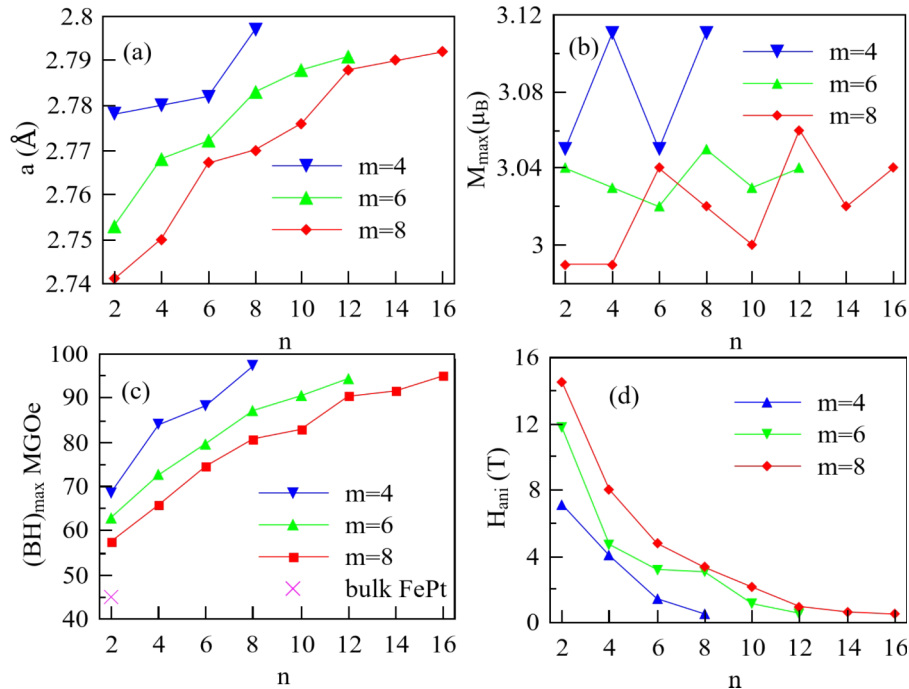


FIG. 2. (a) The in-plane lattice parameter a of the $(\text{FePt})_m/(\text{FeCo})_n$ superlattices. (b) shows the highest local magnetic moment (M_{max}) on Fe atoms. (c) shows the variation of $(\text{BH})_{\text{max}}$ for different m and n . (d) shows the variation of the magnetic anisotropy field, H_{ani} with n (the number of FeCo layers) for different values of m (the number of FePt layers).

towards the bulk value in all cases. Thereafter d_{FeCo} shows only a small variation around 1.46 Å which is close to the bulk value (1.42 Å), but oscillatory behavior of d_{FePt} continues. These results indicate that a *significant anisotropy is created in FeCo by the formation of superlattice*.

Considering the case of the 4-2 superlattice, the optimized in-plane lattice parameters are: $a = b = 2.78 \text{ \AA}$. This is also the nearest neighbor bond length for Fe-Fe as well as Pt-Pt in Fe-Pt layers. This is elongated by 1.95% while the Fe-Fe and Co-Co bonds in FeCo layers are contracted by 2.11% compared to the respective bulk values. This also leads to 2.44% contraction (4.57% expansion) in FePt (FeCo) layer spacing near the interface (Fig. 1) and therefore an increase in the anisotropy in FePt layers while the B2-type FeCo transforms into an anisotropic structure. This is because each atom favors a certain charge density within effective medium theory²⁹ and an expansion (contraction) in FePt (FeCo) in-plane lattice constant is compensated by a decrease (increase) in the interlayer separation of FePt (FeCo) layers near the interface and this further leads to an oscillatory relaxation in other layers as on free surfaces. In 6-2 superlattice, the in-plane Fe-Fe and Pt-Pt bond lengths become smaller ($a = b = 2.75 \text{ \AA}$) whereas d_{FePt} at the interface increases by 1.36% compared with the value for the 4-2 superlattice. The lattice constants $a = b$ increase as n increases (Fig. 2(a)) while d_{FePt} as well as d_{FeCo} decreases as shown in Fig. 1. For the 8-2 superlattice, the in-plane lattice parameters ($a = b = 2.74 \text{ \AA}$) further decrease slightly but the value increases with the number of FeCo layers. In general, the FeCo layers show the largest expansion in d_{FeCo} at the interface and it decreases beyond it and reaches to a saturation value in the middle layer of the FeCo. On the other hand, for FePt layers d_{FePt} contracts the most at the interface and increases beyond it and saturates in the middle. The expansion in d_{FeCo} causes larger average magnetic moment and MAE. Further, the MAE increases with the increase in the number of FePt layers while the magnetization increases with the increase in the number of FeCo layers as FePt is rich in anisotropy and FeCo, in magnetization. This trend is explicit in Fig. 2(c) which shows that the maximum energy product increases with the increase in the number of FeCo layers while it decreases with an increase in the number of FePt layers. Similarly, the anisotropy field increases with the increase in the number of FePt layers while it decreases with the increase in the number of FeCo layers. As m and n vary, the interlayer separations change along with a change in the a lattice parameter and it results in to variation in the hybridization of the orbitals as well as charge transfer leading to an oscillatory behavior in the local highest magnetic moment (Fig. 2(b)).

The calculated average magnetic moments in bulk FePt (FeCo) are 1.62 (2.24) μ_{B} /atom and the local magnetic moments on Fe atoms in bulk FePt (FeCo) from Bader charge analysis are 2.90 (2.74) μ_{B} while the local magnetic moment on Pt (Co) atoms is 0.34 (1.75) μ_{B} . The high magnetic moments on Fe atoms in bulk FePt also arise due to 0.67 e charge transfer from Fe to Pt atoms while in FeCo only 0.16 e charge is transferred to Co atom resulting in a lower value of the magnetic moment on Fe atom. The total magnetic moment per atom for each superlattice is given in Table S1 of supplemental material.³⁰ For a given n , the total magnetic moment per atom decreases with increasing m whereas for a given m it increases to the value of about 2.1 μ_{B} /atom with increasing n . In superlattices the magnetic moments are modified due to structural changes. For the 4-2 superlattice, the average magnetic moment (1.885 μ_{B} /atom) is higher than in bulk FePt. It increases further with FeCo thickness and becomes about 2.10 μ_{B} /atom for 4-8, 4-10, and 4-12 superlattices. This is ~30% higher than the value in bulk FePt. From Bader charge analysis the local magnetic moment at the interface Fe (Fe_{I}) atoms that are nearest to both Co and Pt atoms is 2.87 μ_{B} . These Fe atoms give 0.44 e charge to the neighboring Co and Pt atoms whereas Fe atoms of FePt nearest to the interface ($\text{Fe}_{\text{I-2}}$) have 3.05 μ_{B} magnetic moment in 4-2 superlattice and give 0.68 e to the neighboring Pt atoms (See Fig. 2(b)). The magnetic moments on Fe_{I} ($\text{Fe}_{\text{I-2}}$) atoms are higher than on Fe atoms in bulk FeCo (FePt). The highest value of the magnetic moment in these calculations (3.11 μ_{B} for $\text{Fe}_{\text{I-2}}$ and 2.94 μ_{B} for Fe_{I} atoms) is obtained in 4-8 superlattice. However, beyond the interface region, the magnetic moments decrease and tend to attain the corresponding bulk value. The enhancement in the magnetization in superlattices is attributed to (i) interfacial effects, (ii) tetragonal distortion, (iii) the number of layers of the soft magnet phase, and (iv) slight reduction in the volume. On the other hand, the MAE depends prominently upon the chemical ordering parameter and tetragonal lattice distortion (c/a). In a recent study²⁷ the MAE is reported to be more sensitive to chemical ordering than the tetragonal distortion. For the ordered bulk FePt, the MAE was calculated to be $10 \times 10^7 \text{ erg/cm}^3$ and it shows a decreasing trend

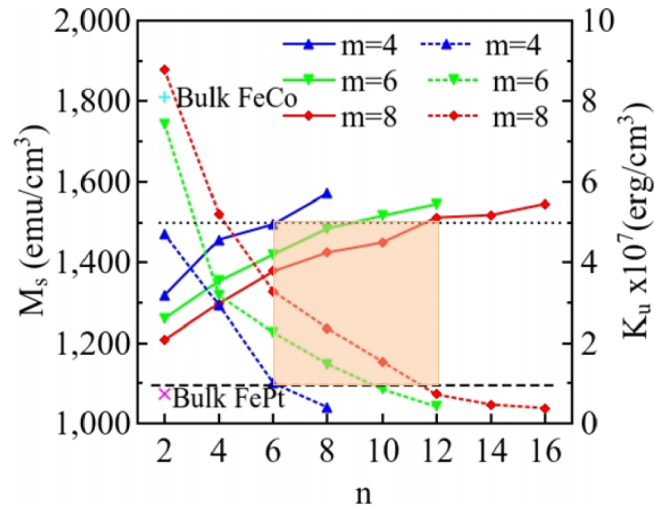


FIG. 3. The saturation magnetization, M_s (left side y axis, solid curves) and magnetic anisotropy energy, K_u (right side y axis, dashed curves) of $(\text{FePt})_m/(\text{FeCo})_n$ superlattices with respect to n (the number of FeCo layers) as m (FePt layers) is varied. The shaded region represents optimal range of n and m values to obtain good permanent magnet behavior. A cross (plus) sign shows the calculated bulk value of the magnetization for FePt (FeCo). The horizontal dashed (dotted) line shows the desirable value of K_u (magnetization) for good permanent magnet behaviour.

with decreasing order parameter. We calculated the MAE to be $12.2 \times 10^7 \text{ erg/cm}^3$ for ordered bulk FePt which is slightly larger (but comparable) as these values are obtained from different methods of calculations and the calculations need high precision. Also in calculations we have perfectly ordered systems but in reality the ordering parameter may be lower and therefore the results of these studies imply a reduction in the value of the MAE for systems with less than perfect order. This could be a possible reason, as stated earlier, for a smaller experimental value of MAE ($6.6 \times 10^7 \text{ erg/cm}^3$) for bulk FePt. However, the order of the MAE is the same in both experiment and calculations.

It is interesting to note here that for the case of $m \gg n$, i.e., for 8-2 superlattice, d_{FePt} tends to approach the bulk FePt value (1.866 \AA) and the anisotropy in FeCo layers is the highest among all the cases we have studied. But the average magnetization (Fig. 3) and therefore $(\text{BH})_{\text{max}}$ (Fig. 2(c)) is low. However, for FeCo rich cases such as the 4-6 superlattice, the anisotropy is on the lower side but it is large enough (Fig. 3) to fulfil the requirement of high magnetization and high enough anisotropy energy for applications. In other cases, such as 6-10 and 8-12 superlattices, there is also reasonably high anisotropy energy as shown in Fig. 3. In order to enhance average magnetization, we need to increase the number of anisotropic FeCo layers. Accordingly, superlattices falling within the shaded region in Fig. 3 are promising for permanent magnets.

The calculated values of M_s , $(\text{BH})_{\text{max}}$, and K_u are listed in Table S2 of supplemental material.³⁰ As shown in Fig. 3, M_s increases with the number of FeCo layers (with lower volume/atom) and decreases with the increase in FePt layers (with higher volume/atom). For 4-2 superlattice the value of M_s is 1318 emu/cm^3 and it reaches the value of 1571 emu/cm^3 for 4-8 superlattice. But in the case of 6-2 (8-2) superlattice it decreases to 1260 (1208) emu/cm^3 . The maximum energy product $(\text{BH})_{\text{max}}$ shows the same trend as M_s and it increases with n (FeCo layers) and decreases with m (FePt layers) as shown in Fig. 2(c). The highest $(\text{BH})_{\text{max}}$ of 97 MGOe is obtained for the 4-8 superlattice among all the systems we have studied. For 6-2 (8-2) superlattice, $(\text{BH})_{\text{max}}$ is calculated to be 63 (57) MGOe. *The value for the 4-8 superlattice is nearly double the calculated value for bulk FePt.*

The value of MAE (K_u) decreases with increasing number of the soft magnet layers as shown in Fig. 3. A large value of K_u is important to develop hard magnets and this puts a limit on the thickness of the soft magnetic phase. We calculated K_u for different FePt/FeCo superlattices and the values are given in Table S2 [Ref. 30]. The value of K_u for the 4-2 superlattice is $4.7 \times 10^7 \text{ erg/cm}^3$ and it decreases (increases) with the increase in the thickness of the soft (hard) magnet phase. A larger value of K_u suppresses the superparamagnetic fluctuations of magnetization and withholds demagnetization. As a

result, it holds the information for longer period. In general, K_u of the order of 10^7 erg/cm³ is desirable. From Table S2 [Ref. 30] and Fig. 3, our calculations show that for superlattices with $m = 4, 6,$ and $8,$ the largest number of FeCo layers is $n = 6, 10,$ and $12,$ respectively, to yield K_u of the order of 10^7 erg/cm³ (see shaded region in Fig. 3). Therefore, we can predict that the optimum thickness of the soft phase is less than ~ 2 nm for the superlattice to work as a good permanent magnet. Also the thickness of the hard magnetic phase should optimally be about 1-2 nm. This would also mean a large number of interfaces which are beneficial for anisotropy and magnetization. We calculated the anisotropy field $H_{\text{ani}} = 2K_u/M_s$ which is the field required to demagnetize the system or flip the magnetization direction. It is found that the suggested thicknesses of the soft and hard phases imply H_{ani} to be about 1 T. Thus, our study suggests the maximum energy product ~ 90 MGOe, $K_u \sim 10^7$ erg/cm³, and anisotropy field $H_{\text{ani}} > 1$ T in these superlattices. Note that from bulk calculations while the MAE is overestimated, the maximum energy product is related to magnetization which is often predicted well from calculations. Even if we increase the cut-off value of K_u to 2×10^7 erg/cm³, we can say that $n = m$ seems to be a good choice. Therefore, our results suggest that a good value of the thickness of the soft magnet phase is around 1-1.5 times the thickness of the hard magnet phase or in the range of 1.5-2.0 nm. Interestingly a similar value of the optimum thickness has been also found for different geometries and materials experimentally.⁴⁻⁶

The orbital magnetic moment L is generally small and it is found to be large along the easy axis in agreement with Bruno model.³¹ As shown in Table S2 [Ref. 30] there is a significant difference in orbital magnetic moment for [100] and [001] directions yielding to large anisotropy. However, spin magnetic moment does not show significant variation along the two mutually perpendicular directions, i.e. it is insensitive to magnetization direction.

IV. CONCLUSIONS

In summary *ab initio* calculations on exchange coupled hard/soft magnet FePt/FeCo superlattices show that the bcc FeCo layers develop anisotropy and it increases with the number of FePt layers. At the interface, the interlayer spacing in FeCo is the largest and it leads to enhancement in the magnetic moments. The average magnetic moments and magnetization increase (decrease) with the increase of FeCo (FePt) layers but the magnetic anisotropy energy decreases as FeCo thickness is increased. Our results suggest that the exchange coupled magnets will have promising properties if the thickness of the soft phase is less than ~ 2 nm. It is to be noted that a similar conclusion has been obtained from recent experiments on FePt and Fe₃O₄ nanoparticle composites as well as on FePt/FeCo thin films. It also implies that the thickness of the hard magnet phase should be of the order of $\sim 1-2$ nm. We believe that our results will have wider applicability such as for nanoparticle composites. We find the highest local magnetic moments on Fe atoms near the interface region. The 4-8 superlattice exhibits the highest magnetic moments as well as saturation magnetization, and thereby the highest maximum energy product (97 MGOe) which is nearly double the calculated value for bulk FePt. The anisotropy magnetic field is ~ 1 T for superlattices with the optimum thickness of the soft and hard phases. For high density recording heads, the desired maximum anisotropy field H_{ani} is 2T. Bulk FePt has large H_{ani} (> 10 T) which precludes its usage as the writing head. This can be reduced by incorporating soft magnet phase such as FeCo as we have shown. Therefore, these spring magnets are also promising candidates for recording media.

ACKNOWLEDGEMENTS

We are grateful to the staff of the Center for Development of Advanced Computing, Pune for allowing the use of their YUVA supercomputing system. We are thankful to AOARD for the financial support under project No. FA2386-13-1-4034.

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