Ab Initio Studies of Magnetic Properties of CoFePd Alloys and Multilayers

Mairbek Chshiev[1] and W. H. Butler[2]

[1]Center for Materials for Information Technology, The University of Alabama, Tuscaloosa, AL 35487 USA
[2]Center for Materials for Information Technology and Department of Physics, The University of Alabama, Tuscaloosa, AL 35487 USA

Enhancement of the saturation magnetization is highly desired for use in the write pole for magnetic recording. There are recent indications that adding small amounts (layers) of Pd to CoFe alloys (multilayers) may enhance their saturation magnetization. In order to investigate this possibility, we have performed *ab initio* studies of the magnetic properties of CoFe–Pd multilayers and alloys. We consistently find that the addition of Pd raises the CoFe sublattice saturation magnetization but simultaneously decreases the total magnetization.

***Index Terms*—High saturation magnetization, magnetic materials, magnetic write heads.**

I. INTRODUCTION

FUNDAMENTAL magnetic properties of transition metals and their compounds continue to attract considerable interest from the scientific community and industry. One property that is simultaneously fundamental, strongly desired, and extremely challenging is a high saturation magnetization (or magnetic flux density B_s). Currently, there is a critical need for higher room temperature B_s for use in write heads in magnetic storage applications. An obvious starting point in the search for high B_s is the FeCo alloy at the maximum value of magnetic moment per atom in the Slater–Pauling curve [1] which is approximately 2.43 T for B_s measured at room temperature for a Co concentration of 30%–40% [2]. Interestingly, these alloys exist in nature in the form of wairauite with about 50% Co concentration [3].

In searching for elements that might enhance the B_s of FeCo, it has been noted that nearly magnetic transition metals such as Pd which are close to a magnetic instability due to an unusually high Stoner enhancement factor [4] can strongly enhance the magnetic moments of dilute Fe [5] or Co [6] impurities. This effect has also been observed in ultra thin Co/Pd and Fe/Pd vapor-deposited multilayers. Saturation magnetization values up to 3.05 T for the Co sublattice in Co–Pd and 2.74 T for Fe sublattice in Fe–Pd at room temperature were observed [7]. A similar observation with B_s for the Fe sublattice being approximately 2.7 T at 15 K was reported for (001)Fe/Pd superlattices prepared by molecular beam epitaxy [8]. It should be noted that these values of saturation magnetization all refer to the Fe or Co sublattice. Based on these results, it has recently been suggested that adding small amounts of Pd to high magnetization FeCo alloys or interspersal of thin layers of Pd in these alloys may further enhance both the CoFe sublattice $B_{s(\mathrm{CoFe})}$ and the total B_s saturation magnetization [9], [10]. In particular, a total B_s of 2.57 T was reported for sputtered (110) Fe70Co30/Pd superlattices [10]. In order to investigate this possibility, we have performed systematic *ab initio* studies of FeCoPd alloys and of multilayers of FeCo and Pd.

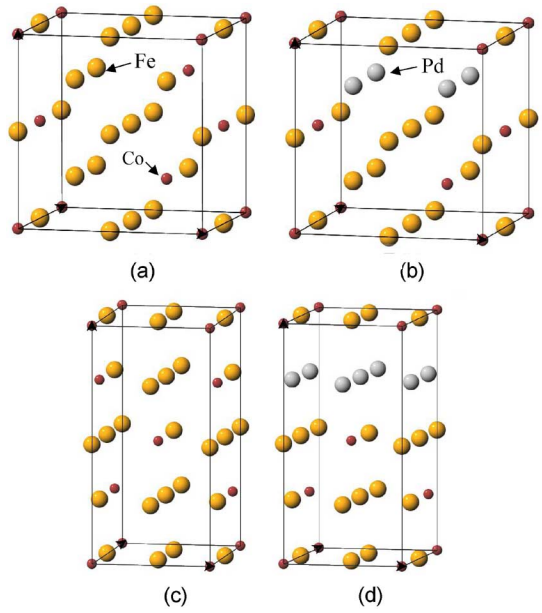

Fig. 1. Sixteen atom cells of $Co_{25}Fe_{75}$ and $Co_{25}Fe_{75}$ Pd in (100) and (110) directions. (a), (b) $Co_{25}Fe_{75}$ and $Co_{25}Fe_{75}$ Pd cells in (100) direction. (c), (d) $Co_{25}Fe_{75}$ and $Co_{25}Fe_{75}$ Pd cells in (110) direction.

II. CALCULATIONS

We began with 16 atom supercells consisting of three atomic layers of $Co_{25}Fe_{75}$ alternating with one atomic layer of Pd in (100) or (110). Thus, both types of cells had four Pd, nine Fe, and three Co atoms as shown in Fig. 1(b) and (d). To show the effect of Pd insertion into these structures, the magnetic moments and magnetizations of these supercells were compared to those of supercells consisting entirely of four layers of $Co_{25}Fe_{75}$ with 12 Fe and four Co atoms in (100) and (110) directions [Fig. 1(a) and (c)]. The calculations were performed using density functional theory (DFT) with the generalized gradient approximation for exchange-correlation potential using the Vienna

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TABLE I
CALCULATED PROPERTIES

direction	atoms/cell	atoms/layer	supercell	volume (Å^3)	CoFe volume (Å^3)	μ(μ_B)	M (10^6 A/m)	B_s (T)	$B_{s(CoFe)}$(T)
(100)	16	4	$[Co_{25}Fe_{75}]_4$	184.94	-	37.43	1.877	2.359	-
	16	4	$[Co_{25}Fe_{75}]_3Pd$	206.17	143.12	31.27	1.407	1.768	2.546
	16	4	$[Co_{25}Fe_{75}]_4$	184.93	-	37.06	1.859	2.335	-
	16	4	$[Co_{25}Fe_{75}]_3Pd$	205.60	148.73	30.59	1.380	1.734	2.397
(110)	48	2	$[Co_{30}Fe_{70}]_{24}$	553.86	-	110.87	1.85	2.33	-
	48	2	$[Co_{30}Fe_{70}]_{23}Pd$	567.86	536.7	108.42	1.77	2.23	2.35
	96	4	$[Co_{30}Fe_{70}]_{24}$	1104.39	-	222.14	1.865	2.344	-
	96	4	$[Co_{30}Fe_{70}]_{23}Pd$	1124.76	1064.39	214.51	1.869	2.222	2.349

Calculated properties of relaxed $[CoFe]_n$ and $[CoFe]_nPd$ supercells in (100) and (110) direction: volume after relaxation, total magnetic moment μ, magnetization M, total and CoFe sublattice magneticflux density (saturation magnetization) B_s and $B_{s(CoFe)}$. All $[CoFe]_nPd$ structures have only one Pd layer and n indicates number of CoFe layers.

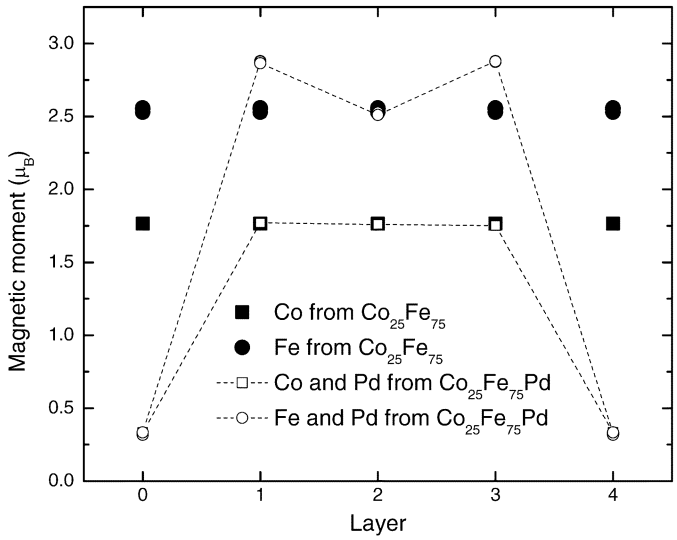

Fig. 2. Layer distributions of local Co (squares) and Fe (circles) atomic magnetic moments in $Co_{25}Fe_{75}$ and $Co_{25}Fe_{75}$ Pd (100) cells represented in Fig. 1(a) and (b). CoFe layer replacement with Pd corresponds to layers 0 and 4, which are the same due to periodicity of the structure.

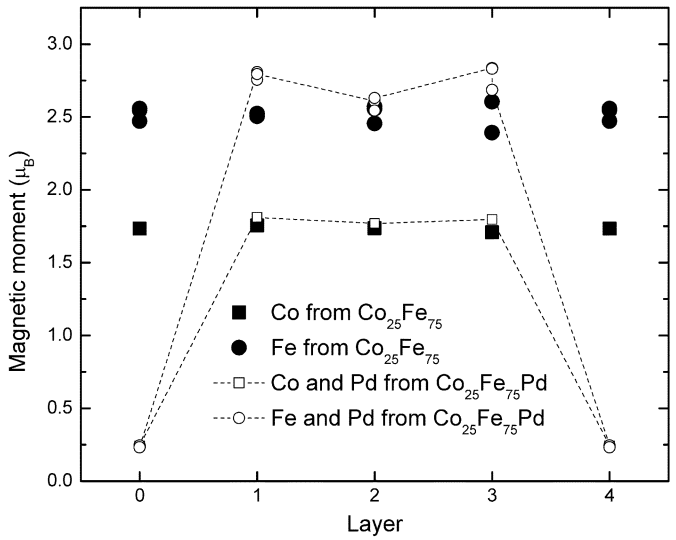

Fig. 3. Same as in Fig. 2 for (110) cells represented in Fig. 1(c) and (d).

ab initio simulation package (VASP) [11]–[14] with PAW pseudopotentials [15], [16]. All supercells were completely relaxed structurally.

For the $Co_{25}Fe_{75}$ structure in the (100) direction [Fig. 1(a)], we found the total magnetic moment to be 37.43 μ_B after structural and volume relaxation which gave a magnetization of 2.36 T. When one CoFe layer was replaced with Pd, i.e., for the $Co_{25}Fe_{75}$ Pd in the (100) direction Fig. 1(b), the total magnetic moment dropped to 31.27 μ_B after relaxation giving a magnetization 1.77 T. However, the CoFe "sublattice magnetization" in the $Co_{25}Fe_{75}$ Pd cell calculated by taking into account the fraction of cell volume occupied by the CoFe sublattice after relaxation was equal to 2.55 T. For $Co_{25}Fe_{75}$ and $Co_{25}Fe_{75}$ Pd in the (110) direction shown in Fig. 1(c) and (d), we obtained magnetic moments of 37.06 and 30.59 μ_B, respectively, corresponding to saturation magnetization values of 2.34 and 1.73 T. The CoFe sublattice saturation magnetization for the supercell with the Pd layer was found to be 2.34 T. These results are summarized in the first four lines of Table I.

We have also found that the Pd layer induced enhanced magnetic moments in neighboring Fe atoms consistent with previous reports for Fe/Pd multilayers [17]. This is shown in Figs. 2 and 3 for the (100) and (110) structures, respectively. For (100) with no Pd layer presence [Fig. 1(a)] the moments are distributed almost uniformly across the structure (see Fig. 2) with about 1.77 μ_B for Co (solid squares) and 2.55 μ_B for Fe (solid circles) atoms. The situation changes when one CoFe layer is replaced with Pd Fig. 1(b). In this case, the magnetic moments of the Fe atoms neighboring the Pd layer are enhanced up to 2.87 μ_B (open circles in Fig. 2). A similar picture holds for the case of (110) structures (Fig. 3). There is a small dispersion in the moments due to the random distribution of Co atoms from layer to layer in our model of the CoFe alloy.

We have also investigated much larger cells consisting of 24 atomic layers in the (110) direction. Two- and four-atom per layer cases were considered giving supercells with a total of 48 atoms (14 Co, 32 Fe, and two Pd) or 96 atoms (28 Co, 64 Fe, and four Pd) representing 23 layers of $Co_{30}Fe_{70}$ and one Pd layer. These cells were designed to model the results of [10] and were again compared with similar cells that were entirely CoFe. After structural and volume relaxation, the magnetization of the $[CoFe]_{23}Pd_1$ supercell in both cases was found to be 2.22–2.23 T compared to 2.33–2.34 T for the $[CoFe]_{24}$ cell. The deduced CoFe sublattice saturation magnetization was equal to 2.35 T. These results are summarized in the last four lines of Table I.

III. Conclusion

The results represented above suggest that within the approximations of our calculations, the addition of Pd raises the FeCo sublattice magnetization but decreases total magnetization. We consider the most important approximations in these calculations to be: 1) the use of the generalized gradient approximation (GGA) to density functional theory (DFT) and 2) neglect of the spin-orbit interaction and orbital moments.

Concerning 1), our experience has been that DFT in the GGA approximation is usually reliable in predicting spin magnetic moments for transition metals and compounds. Often, the magnetic moment will be given correctly even when other properties are not. One caution, however, is that the magnetic moment of atoms such as Pd which are very near a nonmagnetic-to-magnetic transition may be very sensitive to volume and to other details of the calculation such as the exchange-correlation functional.

Concerning 2), our neglect of orbital contributions to the magnetization undoubtedly decreased the magnetization of both the FeCo supercells and the supercells containing Pd. Magnetomechanical measurements of the electron g-factor [18] indicate that an enhancement of the magnetization of about 4%–5% should be expected from orbital effects.

In our opinion, it is unlikely that orbital effects would be sufficient to overcome the deficit in spin moment produced by the addition of the Pd atoms. When an Fe or Co atom is replaced by a Pd atom, the moments of neighboring Fe atoms may be increased by as much as 0.32 μ_B and the Pd atom attains a moment of approximately 0.3 μ_B, this is unlikely to be enough to offset the loss of the moment of the Fe or Co atom replaced and the increase in volume which is approximately 0.44% for every 1% of FeCo replaced by Pd. It should be noted that on very general grounds, the spin moment of Pd is limited to approximately 0.6 μ_B by the filling of the d-shell. Similarly, the spin moment of Fe is limited to about 3 μ_B (assuming 7 d-electrons) and that of Co to about 2 μ_B (assuming 8 d-electrons).

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