Giant Magnetic Anisotropy in Tetragonal FeCo Alloys

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In order to further increase the recording density in hard disk drives, new media materials are required. Two essential parameters of future recording media are a large uniaxial magnetic anisotropy energy (MAE) K_u and a large saturation magnetization M_s. Based on first-principles theory, we predict that very specific structural distortions of FeCo alloys possess these desired properties. The discovered alloy has a saturation magnetization that is about 50% larger than that of FePt—a compound that has received considerable attention lately—with a uniaxial MAE that can easily be tailored reaching a maximum value that is 50% larger than that of FePt.

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The recording density of hard disk drives has increased by more than 6 orders of magnitude since the first commercial product was released about 50 years ago. This remarkable increase in the areal density, that is, the amount of information that can be stored per unit area, has been achieved mainly by a simple scaling of the dimensions of the bits recorded in the storage layer [1]. This traditional scaling, if extrapolated, will soon reach the superparamagnetic limit, i.e., the ratio of the magnetic energy per grain $K_u V$, where V is the grain volume, to the thermal energy $k_B T$ [1,2], becomes sufficiently small that recorded data will be erased by thermal fluctuations in an intolerable short time. In order to allow for a further decrease of the grain volume in future recording media while maintaining the integrity of the recorded data, high K_u materials [3] are needed. However, the maximum practical magnetic anisotropy energy (MAE) is limited by the required write field $H_w \approx K_u/M_s$ that must be delivered by the writer. This, in turn, is limited by the writer's saturation magnetization and its geometry. Thus, a large saturation magnetization M_s is an essential feature of materials for recording media. In addition, the amplitude of the magnetic field emanating from recorded data scales with the product of M_s and the thickness of the recording layer. Hence, a larger M_s increases the field available at the read-back process. Currently, much attention is being paid to chemically ordered FePt, a compound that indeed possesses a large MAE and a large saturation magnetization [3]. The use of FePt in practical applications as a recording layer is complicated by the fact that a phase transition to the ordered $L1_0$ phase has to be induced at a relatively high temperature [4–6], which increases diffusion of other atomic species used in the recording disk. From first-principles calculations, we predict that tetragonal FeCo alloys of a very specific structure and alloy concentration have much improved properties for future recording media, i.e., a giant MAE and a large saturation magnetic moment.

Because of the simplicity in the synthesis and the abundance of material, magnetic applications frequently make use of Fe-based alloys. If one is interested in high moment materials, it is often FeCo alloys that are under focus. At the cubic structures, the MAE of Fe and the other ferromagnetic transition metals is far too low to be of interest as recording media, as it is of the order of μeV/atom. However, if the cubic symmetry is broken, the MAE increases by several orders of magnitude. As we shall see below, for very specific choices of structural distortion and an optimized valence electron concentration, an FeCo alloy with a giant MAE and a large saturation magnetic moment is identified that has great potential for use in magnetic recording media.

In the following, we present first-principles calculations of the uniaxial MAE of tetragonal $Fe_{1-x}Co_x$ within the whole concentration range. The MAE is calculated as $K_u = E^{100} - E^{001}$, where E^{100} and E^{001} are the total energies with the magnetization in the [100] and [001] directions, i.e., perpendicular or parallel to the c axis, respectively. All results presented here were obtained with a fully relativistic implementation of the full-potential linear muffin tin orbitals (FP-LMTO) method [7] and the force theorem [8,9]. The local density approximation was used for the exchange correlation potential, and the integration in reciprocal space was done with approximately 6.5×10^4 **k** points in the full Brillouin zone using the modified tetrahedron method [10]. For more details about the calculations, see Ref. [11]. The results for the disordered alloys that are presented here were obtained with the virtual crystal approximation (VCA), which was demonstrated to work well for the ferromagnetic transition metals [12]. To check the applicability of the VCA, the MAE of ordered $Fe_{0.5}Co_{0.5}$ in the tetragonally distorted CsCl structure was calculated for some selected c/a ratios and compared to the corresponding VCA results. The qualitative behavior of the MAE is similar, but small quantitative differences arise due to the different state of order considered for the alloys.

In Fig. 1 (upper plot), the uniaxial MAE of tetragonal $Fe_{1-x}Co_x$ is shown as a function of the c/a ratio and the Co concentration x [13]. Within a certain region of the phase space covered by our calculations, around a c/a ratio of 1.20–1.25 and 60% Co, the MAE increases enormously in magnitude and reaches a value of the order of 700–800 μeV/atom. The MAE per volume, which is the technologically relevant figure, is thereby estimated to be more than 50% larger than the largest experimental result for FePt reported thus far [14,15]. Note that the magnetic easy axis is oriented along the [001] direction so that the alloy can be used in perpendicular magnetic recording applications. It is important to point out that the agreement between calculated and experimental values for the MAE of ferromagnetic transition metals, compounds, surfaces, and superlattices is in general satisfactory, and is known to be very accurate for strained Fe, Co, and Ni [11,16–21]. It is clear from Fig. 1 that only for very specific concentrations and c/a ratios does the alloy exhibit these large values of the MAE, something we will return to below. In the lower plot of Fig. 1, the saturation magnetic moment of the FeCo alloys is shown. This function has its maximum at a Co concentration around 20%, as is the case for bcc FeCo alloys [22]. However, for the concentrations where the uniaxial MAE is very large, the magnetic moments are of the order of $2.1\mu_B$, which results in a more than 50% larger saturation magnetization than that of the FePt compound.

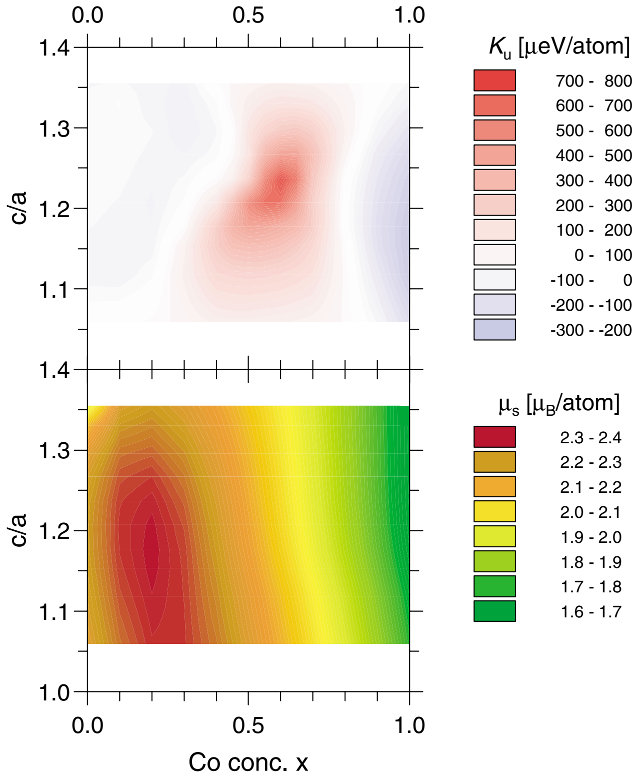

FIG. 1 (color). Calculated uniaxial MAE K_u (upper panel) and saturation magnetic moment μ_s (lower panel) of tetragonal $Fe_{1-x}Co_x$ as a function of the c/a ratio and the Co concentration x.

In order to compare the calculated MAE and saturation magnetic moment of the FeCo alloys shown in Fig. 1 to other high K_u materials, e.g., chemically ordered FePt, we have in Fig. 2 reproduced the stability contour plot from Ref. [3] and included our result for the FeCo alloy that yields the maximum uniaxial MAE. The stability boundary (dotted line) in Fig. 2 was obtained from micromagnetic calculations [23] and represents the optimal media design in terms of saturation magnetization and MAE. Media that are placed below the boundary have grains that are thermally stable but too large for high density recording. Media above the stability boundary allow for smaller grains and henceforth high density recording, but unfortunately they require a write field that is too large. As seen from the figure, the FeCo alloy proposed here has characteristics that are superior to the media materials that are currently in use or under consideration for high density recording media. The strength of the FeCo alloys is the much larger saturation magnetization and the ability to tailor the uniaxial MAE within a wide range by changing the alloy concentration (cf. Fig. 1). Note that the stability condition shown in Fig. 2 was based on parameters that are somewhat outdated, and the boundary line should be moved upward in the figure to account for parameters of current devices. However, the figure serves well to illustrate our main point that the proposed FeCo alloy is superior even to FePt from a magnetic point of view.

In order to understand why the MAE becomes large only at very specific values of the Co concentration and

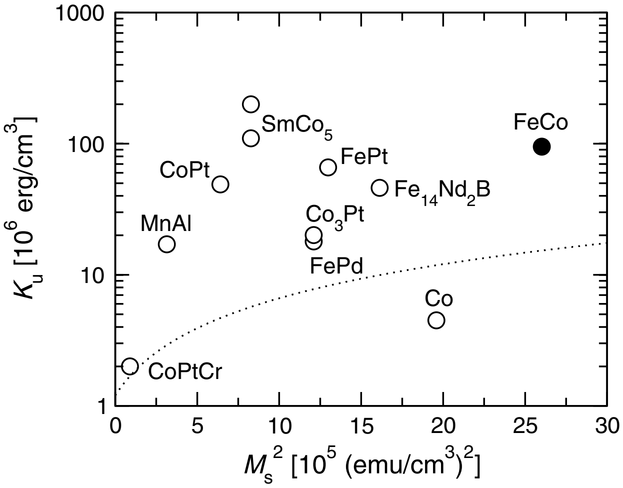

FIG. 2. Stability contour plot of high K_u materials, drawn after Fig. 1 of Ref. [3] (open circles). The present result for the tetragonal FeCo alloy with the maximum uniaxial MAE is included for comparison (closed circle). The dotted line is the 40 Gbits/in^2 stability boundary according to Charap *et al.* [23], assuming a write field of 5100 Oe and 12 nm grains.

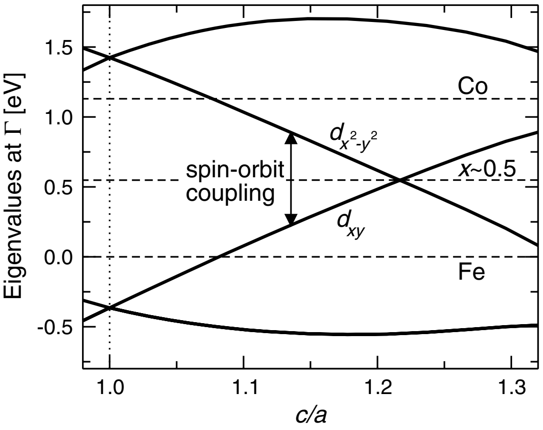

FIG. 3. Calculated d dominated spin-down eigenvalues at the Γ point of tetragonal Fe relative to the Fermi energy as a function of the c/a ratio. The energy scale is chosen so that E_F is at zero for Fe. The Fermi energies of Fe, Co, and $Fe_{1-x}Co_x$ with $x \approx 0.5$ are indicated by dashed lines.

c/a ratios, we proceed with an analysis based on the electronic structure. In Fig. 3, we show the d dominated eigenvalues of the spin-down band of bcc Fe at the high-symmetry Γ point. Upon a tetragonal distortion, the triply degenerate t_{2g}-like eigenvalues and the doubly degenerate e_g-like eigenvalues are split. The t_{2g}-like eigenvalue with the orbital character d_{xy} moves upward in energy while the e_g-like eigenvalue with $d_{x^2-y^2}$ orbital character moves downward. In the fcc limit, i.e., at $c/a = \sqrt{2}$, the $d_{x^2-y^2}$ state of the bcc structure becomes a d_{xy} state, and vice versa, due to a 45° rotation of the coordinate system between these two structures. It is known that the microscopical mechanism behind the MAE is the spin-orbit interaction [24,25], and since it is quite small in the ferromagnetic transition metals one can use perturbation theory. It can be shown that, in second order perturbation theory, the MAE is caused by matrix elements of the spin-orbit interaction between occupied and unoccupied states, as illustrated for the d_{xy} and $d_{x^2-y^2}$ states in Fig. 3. Since the MAE in this theory is inversely proportional to the energy difference between the occupied and unoccupied states, a strongly enhanced MAE can be expected for tetragonal FeCo alloys, when the c/a ratio is close to 1.22 and the valence electron configuration is such that the Fermi energy is located where the d_{xy} and $d_{x^2-y^2}$ states cross. In a rigid band picture, the alloy concentration that should result in the maximum MAE can be estimated to be around 50%, which is in satisfactory agreement with the concentration obtained from Fig. 1. Our analysis is, of course, general and applies to all states in the Brillouin zone. Hence, tuning the MAE only by inspection of alloy conditions where d_{xy} and $d_{x^2-y^2}$ cross relies on the fact that other contributions in reciprocal space are less important, something which for Fe has been shown to hold [11,21].

A possible route to manufacture tetragonally distorted FeCo alloys is by epitaxial growth on a suitable substrate, or as a building block of a superlattice, such that the desired c/a ratio is obtained. That distorted structures of this kind are possible to accomplish, albeit with c/a ratios that are slightly smaller than suggested here, was exemplified in numerous studies. If thin films of Fe [26] and Co [27] are grown on Rh(100), c/a ratios of 1.16 and 1.19, respectively, are obtained. Using Pd(100) as the substrate, the corresponding numbers are 1.11 for Fe [28] and 1.15 for Co [29]. Engel and co-workers [30] manufactured a (001) oriented Co/Pd superlattice with an estimated c/a ratio of 1.19. Changing the relative amount of the constituents in such a superlattice allows one to change the c/a ratio, and, hence, the MAE, within a certain range. If the magnetic Co layer is replaced by an FeCo alloy, the alloy concentration emerges as an additional parameter to tailor the MAE within a wide range. Hence, it should be possible to find the optimum alloy condition and structural distortion to fabricate FeCo alloys with highly improved characteristics for use as recording materials. Also, a further enhancement of the MAE can be expected due to the strong spin-orbit coupling of a heavy ligand atom [31], such as Pd or Pt, in a superlattice. In that way, a large MAE can be obtained even at a nonideal c/a ratio. Thus, the presently suggested FeCo alloys grown in a superlattice geometry together with Pt should be very promising. We note finally that the considerably lower deposition temperature of FeCo alloys [32], as compared to that of the chemically ordered phase of FePt [4–6], is of great technological importance.

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