Unidirectional anisotropy in ultrathin transition-metal films

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Unidirectional magnetic anisotropies in low-symmetry magnetic thin films such as cobalt on vicinal copper surfaces are investigated. Possible explanations of the observed uniaxial anisotropies are competing anisotropy (CA) coefficients and Dzyaloshinskii-Moriya (DM) interactions. Unidirectional CA is an interesting mechanism occurring in low-symmetry magnets and involves neither antiferromagnetic exchange nor spin canting. It is visible in the easy-cone regime and decides, for example, whether the preferential magnetization direction points up or down a stepped surface. In the case of Co/Cu(111) films, however, the anisotropy direction speaks in favor of DM-type interactions. [S0163-1829(98)03541-3]

I. INTRODUCTION

The phenomenon of magnetic anisotropy and its atomic explanation is one of the most compelling subjects in solid-state and surface science. Magnetic anisotropy means that the energy of a magnet contains an anisotropic contribution \( E_a(M) = E_a(-M) \), where \( M \) is the magnetization.\(^{1,2} \) In most cases, this anisotropy gives rise to symmetric hysteresis loops \( M(-H) = -M(H) \), where \( H \) is the external field and \( M \) is the average magnetization in the field direction (dashed line in Fig. 1). A unidirectional shift of the hysteresis loop (solid line in Fig. 1) is observed, for example, in exchange-anisotropic magnets such as cobalt particles coated by cobaltous oxide CoO.\(^{3,4} \) A similar effect is caused by Dzyaloshinskii-Moriya (DM) interactions,\(^{5-7} \) which occur in metallic spin-glasses and in low-symmetry insulators such as \( \alpha \)-Fe\( _2 \)O\( _3 \).\(^{8-10} \) In the context of ultrathin-film magnetism, unidirectional Kerr hysteresis loops have been observed for stepped Co surfaces: Co/Cu(1117) films vicinal to fcc (001) exhibit an unambiguous but unexplained unidirectional shift of order 1 mT.\(^{11,12} \) Although nonferromagnetic spin structures cannot be ruled out in low-symmetry 3d films,\(^{13} \) it is not possible to ascribe the observed loops to the well-known exchange between ferromagnetic and antiferromagnetic phases as in Co/CuO.

The aim of this paper is to draw attention to the phenomenon of unidirectional anisotropies in low-symmetry transition-metal films and to discuss possible physical explanations. In particular, we will discuss under which conditions competing anisotropy (CA) contributions yield unidirectional hysteresis loops.

II. COMPETING ANISOTROPIES

Consider, for the moment, uniformly magnetized films characterized by the magnetization direction \( \mathbf{n} = \cos \theta \mathbf{e}_z + \sin \theta \cos \phi \mathbf{e}_x + \sin \theta \sin \phi \mathbf{e}_y \), where it is common to write the magnetic anisotropy energy in terms of expressions such as\(^{14-16} \)

\[
E_a(\theta) = K_1 \sin^2 \theta + K'_1 \sin^2 \theta \cos(2 \phi - 2 \phi_0) \\
+ K_2 \sin^4 \theta + \cdots.
\]

Here \( K_1 \) and \( K_2 \) are the first and second uniaxial anisotropy constants, respectively, and \( K'_1 \) describes deviations from the uniaxial anisotropy. The disadvantage of the functions established by Eq. (1) is that they are neither complete nor orthogonal. The nonorthogonality means, for example, that \( K_1 \) is not a true lowest-order anisotropy constant but contains an admixture of higher-order atomic contributions.\(^{17,18} \) In fact, the validity of Eq. (1) is limited to highly symmetric structures, such as low-indexed bcc and fcc surfaces.

A complete and orthonormal set of functions is obtained by using Legendre polynomials.\(^{17,18} \) For example, neglecting higher-order and nonuniaxial contributions we reproduce the well-known expression

\[
E_a = \frac{K_1}{2} (3 \cos^2 \theta - 1) + \frac{K_2}{8} (35 \cos^4 \theta - 30 \cos^2 \theta + 3),
\]

where the \( K_n \)’s are the \( n \)th order uniaxial anisotropy coefficients. Minimizing Eq. (2) with respect to \( \theta \) yields the phase diagram Fig. 2(b). Note that the uniaxial relations \( K_1 = -3K_2/2 - 5K_4 \) and \( K_2 = 35K_4/8 \) transform Fig. 2(b) into the more familiar diagram Fig. 2(a). We will see that the easy-cone regime, characterized by an angle \( \theta_e = \arcsin[|K_1|/K_2] \) between the \( z \) axis and \( \mathbf{n} \), is of particular importance in the present context. It is worth emphasizing that the “leading” anisotropy constant \( K_1 \) is an effective parameter which may be very small due to demagnetizing-field contributions of order \( -\mu_0 M^2 \).\(^{12,14} \)

FIG. 1. Uniaxial and unidirectional hysteresis loops (schematic). \( M \) is the average magnetization direction parallel to the field \( H \).
The sin^2 f M from also in papers dealing with nonideal surfaces. \textsuperscript{19} general reviews on thin-film and surface anisotropies but publishes two equivalent cones at \( u \bar{u} \) barriers at ever, Fig. 2 shows that the two cones are separated by energy limit, where \( E_a(\theta) = E_a(\pi - \theta) \).

Adding the lowest-order nonuniaxial contributions to Eq. (2) yields the anisotropy energy

\[
E_a = E_u + \kappa_{21}\cos \theta \sin \theta \cos \phi + \kappa_{2s}\sin^2 \theta \cos 2\phi + \kappa_{2s}\sin^2 \theta \sin 2\phi, \tag{3}
\]

where \( \kappa_{2mc} \) and \( \kappa_{2ms} \) are nonuniaxial anisotropy coefficients. The \( \sin 2\phi \) and \( \cos 2\phi \) terms are related to the \( \cos(2\phi - 2\phi_0) \) term in Eq. (2): \( 2\phi_0 = \arctan(k_{22x}/k_{22z}) \) and \( K'_{2x} = \sqrt{k_{22x}^2 + k_{22z}^2} \). Here we are interested in the unidirectional coefficients \( \kappa_{21c} \) and \( \kappa_{21s} \), which are ignored not only in general reviews on thin-film and surface anisotropies but also in papers dealing with nonideal surfaces.\textsuperscript{19}

From the prefactor \( \sin \theta \cos \theta \) we deduce that there is no unidirectional anisotropy for \( \theta = 0 \) and \( \theta = \pi/2 \). In the intermediate easy-cone regime, \( 0 < \theta < \pi/2 \), the magnetization prefers some unique in-plane angle \( \phi \). It is important to keep in mind that Eq. (3) does not break the global inversion symmetry \( E_a(-\mathbf{M}) = E_a(\mathbf{M}) \), which is realized by simultaneously changing \( \phi \rightarrow -\phi + \pi \) and \( \theta \rightarrow \pi - \theta \). In the uniaxial limit, where \( E_a \) is independent of \( \phi \), this symmetry establishes two equivalent cones at \( \theta = \theta_c \) and \( \theta = \pi - \theta_c \). However, Fig. 2 shows that the two cones are separated by energy barriers at \( \theta = 0 \) and \( \theta = \pi/2 \), so that intercone transitions from \( \mathbf{M} \) to \( -\mathbf{M} \) require comparatively large activation energies. The external magnetic field necessary to overcome the intercone barrier depends not only on the anisotropy coefficients of the film but also on the field direction. However, in nearly ideal films it is much larger than the unidirectional shift of the hysteresis loop. It is worthwhile noting that a similar energy barrier exists for Co/CoO-type unidirectional anisotropies. In that case, the field necessary to overcome the barrier is given by the antiferromagnetic CoO exchange field. Figure 3 shows how the low-symmetry anisotropy contributions contained in Eq. (3) perturb uniaxial energy landscapes. For the practical realization of the energy landscapes Fig. 3 see Sec. III.

To estimate \( \kappa_{12c} \) and \( \kappa_{12s} \) we start from Néel’s pair anisotropy energy \( g(3\cos^2\alpha - 1)/2 \), where \( \alpha \) is the angle between \( \mathbf{n} \) and the real-space vector \( r_i = r_j \) connecting the positions of two nearest neighbors, and \( g \) is a phenomenological coupling constant.\textsuperscript{1} Note that the applicability of the Néel model to itinerant magnetism is only semiquantitative but gives a good account of the symmetry aspect of the problem.\textsuperscript{20} The spherical harmonic addition theorem\textsuperscript{21} yields, after straightforward calculation, the single-atom coefficients

\[
\kappa_{21c} = \frac{g}{2} \sum \sin \Theta_i \cos \Phi_i \cos \phi_i, \tag{4a}
\]

\[
\kappa_{21s} = \frac{g}{2} \sum \sin \Theta_i \cos \Phi_i \sin \phi_i, \tag{4b}
\]

where \( \Theta_i \) and \( \Phi_i \) describe the relative position of the \( i \)th neighbor. The total anisotropy is obtained by adding all atomic contributions.

### III. Co/Cu(111) SURFACES

It is interesting to compare the predictions Eq. (4) with the behavior of fcc \((11n)\) surfaces vicinal to fcc \((001)\).\textsuperscript{11,12,22–24} Figure 4 shows the atomic structure of fcc \((11n)\) surfaces, which involve four types of atoms: bulk atoms, surface atoms, step-edge atoms, and step-corner atoms.\textsuperscript{22} Since there are no second-order bulk contributions, we have to deal with \( N_s \) surface atoms, \( N_c \) step-edge atoms, and \( N_e \) step-corner atoms. Up to higher-order terms, the summation procedure yields the CA energy...
where $k$ indicates that the last term in Eq. mechanism, the fictitious unidirectional anisotropy field $D$ anisotropy-field contribution. Independently of the hysteresis yields a displacement of the hysteresis loop $u$ multiplied by the prefactor $\sin$.

A Zeeman-energy contribution proportional to $\sin(\mu H)$ magnetic field applied perpendicular to the step edges yields a shift in the hysteresis loop. This is seen as a Zeeman effect can be expected for stepped surfaces, because surface, step-edge, and step corner atoms have different electronic properties local density of states and moments. From the symmetry of the fcc $(11n)$ surfaces (Fig. 4) follows that the step-corner atoms yield a nonzero DM anisotropy parallel to the step edges, which is in agreement with experiment.

A particular point about the DM anisotropy is that the atomic spins enter the interaction as $S_i \times S_j$, so that they have to be noncollinear to yield unidirectional anisotropy. At this stage it remains open whether this noncollinearity reflects parasitic spin canting as in $\alpha$-Fe$_2$O$_3$ (Ref. 10) or micromagnetic deviations from the ideal spin alignment. In any case, we are convinced that this work will stimulate further experimental and theoretical research in the field of unidirectional anisotropies in low-symmetry ultrathin transition-metal films. This refers not only to the atomic and micromagnetic spin structures but also to problems such as intercone transitions in external fields.

In conclusion, we have established and analyzed the existence of unidirectional anisotropies in stepped ultrathin transition-metal films. In the case of Co/Cu$(11n)$, the unidirectional hysteresis loops are ascribed to Dzyaloshinskii-Moriya-type interactions, but in general there is a possibility of a unidirectional anisotropy associated with competing anisotropy coefficients. This anisotropy, which has not been considered in previous work, is nonzero for low-symmetry easy-cone configurations and involves neither DM nor antiferromagnetic exchange.

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19 Note that second-order unidirectional contributions can be eliminated by a unitary transformation, but this “simplification” is paid by the introduction of a fictitious reference frame generally unrelated to directions such as [001] or [11n]. Furthermore, the transformation procedure creates higher-order low-symmetry anisotropy expressions.