



# Magnetic anisotropy from single atoms to large monodomain islands of Co/Pt(111)

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## Abstract

By studying the magnetic behavior of self-assembled Co islands on a single-crystal metal surface, Pt(111), we show how the magnetic anisotropy evolves from isolated atoms to monolayer islands and films. Single Co adatoms are found to have a giant magnetocrystalline anisotropy energy of  $E_a = 9.3 \pm 1.6$  meV/atom arising from the combination of partly preserved orbital moments ( $m_L = 1.1 \mu_B$ ) and strong spin-orbit coupling induced by the Pt substrate. Combined scanning tunneling microscopy and X-ray magnetic circular dichroism experiments performed for differently sized small two-dimensional Co islands establish a clear connection between  $E_a$  and  $m_L$ , both quantities decrease sharply with the lateral coordination of the magnetic atoms. In accordance with this, Kerr magnetometry experiments, again performed in conjunction with scanning tunneling microscopy, reveal that the anisotropy energy of large two-dimensional Co islands is almost entirely determined by the relatively low number of perimeter atoms, having  $E_a = 0.9 \pm 0.1$  meV/atom. These results confirm theoretical predictions and are of fundamental value the understanding of how the magnetic anisotropy develops in finite-size magnetic particles. Identification of the role of perimeter and surface atoms opens up new opportunities for engineering the anisotropy and the moment of a magnetic nanostructure. **To cite this article:** *P. Gambardella et al., C. R. Physique 6 (2005).*

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## Résumé

**Anisotropie magnétique de l'atome isolé aux îlots monocouches de Co/Pt(111).** En étudiant les propriétés magnétiques de particules de Co auto assemblées sur une surface d'orientation (111) d'un monocristal de Pt, nous montrons comment l'anisotropie magnétique évolue de l'atome isolé aux îlots monocouches et aux films ultramincés. Nous trouvons que les atomes de Co isolés adsorbés en surface ont une énergie d'anisotropie magnétocrystalline de  $E_a = 9.3 \pm 1.6$  meV/atome provenant de la combinaison d'un moment orbital conservé ( $m_L = 1.1 \mu_B$ ) et d'un fort couplage spin-orbite induit par le substrat de Pt. Des expériences combinées de microscopie à effet tunnel et de dichroïsme circulaire magnétique à rayons X, effectués sur des îlots de Co de petit taille établissent une connexion claire entre  $E_a$  et  $m_L$ , chacune de ces quantités décroissant rapidement avec la coordination latérale des atomes magnétiques. En conformité avec ceci, nous trouvons en employant la magnétométrie par effet Kerr et le microscope à effet tunnel que l'énergie d'anisotropie de grands îlots bidimensionnels de Co est presque entièrement déterminée par le nombre relativement faible d'atomes au périmètre, ayant  $E_a = 0.9 \pm 0.1$  meV/atome. Ces résultats confirment les prédictions théoriques et sont d'un intérêt fondamental pour comprendre comment l'anisotropie magnétique se développe dans les particules magnétiques de taille finie. L'identification du rôle des atomes au périmètre et des atomes de surface ouvre de nouvelles opportunités pour l'ingénierie de l'anisotropie et du moment de nanostructures magnétiques. **Pour citer cet article :** *P. Gambardella et al., C. R. Physique 6 (2005).*

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## 1. Introduction

It is well known that the magnetic properties of transition metal particles depend on the coordination of the constituent atoms [1–4]. Small clusters deposited on metal surfaces are predicted to have spin ( $m_S$ ) and orbital ( $m_L$ ) magnetic moments in between those of bulk compounds and free atoms [5–7], and to exhibit strong magnetic anisotropy with characteristic energies ( $E_a$ ) of the order of 1–10 meV/atom, i.e., a factor  $10^3$  larger than bulk ferromagnetic metals [7,8]. In the case of single-domain particles,  $E_a$  determines the orientation and stability of the magnetization and is thus a crucial parameter for most applications of magnetic materials in modern technology. Van Vleck [9] was among the first to recognize that the orbital magnetization, via the spin-orbit interaction, connects the magnetocrystalline anisotropy to the atomic structure of magnetic materials.

In recent years, extensive work [10–19] on magnetic surfaces and thin films has shown that a lowering of the symmetry results in an increase of  $m_L$  compared to bulk systems, where the  $d$ -state hybridization and the crystal field effectively quench  $m_L$ . This effect gives rise to a variety of interesting phenomena, such as enhanced and perpendicular magnetic anisotropy [2,12–16, 19]. Surface-supported nanoparticles offer additional degrees of freedom to tune the magnetic anisotropy by ad-hoc modifications of the particle size, shape, and coupling with the substrate, making nanometer sized systems attractive for basic investigations as well as for miniaturized data storage applications [20–22]. In this respect, self-assembled magnetic clusters grown on metal surfaces constitute an ideal system since their size, shape, and composition can be controlled with relative ease while producing them in numbers large enough for investigation with spatially integrating techniques [23,24]. In the following we present an extensive study of the evolution of the magnetic anisotropy energy in surface supported aggregates when their sizes evolve from one to about one thousand atoms. The data demonstrate a strong correlation between magnetic properties and morphological details. Reducing the atomic coordination number the anisotropy energy is seen to steeply increase and it assumes the giant value of 9 meV/atom in the case of a single adatom. Identification and evaluation of the role of high and low coordinated magnetic atoms opens up new opportunities for material engineering which are likely to be of help in pushing forward the superparamagnetic limit in small magnetic particles.

## 2. Orbital magnetic moment and magnetic anisotropy of single Co atoms and nanoparticles

Gas-phase transition metal (TM) atoms possess large  $m_S$  and  $m_L$  according to Hund's rules, which are due to intra-atomic Coulomb interactions. In a solid, electron delocalization and crystal field effects compete with these interactions, causing a substantial decrease of  $m_S$  and partial or total quenching of  $m_L$ . Theoretical calculations [7] predict such effects to be strongly reduced in TM impurities at non-magnetic surfaces owing to the decreased coordination, with implications also for the appearance of significant magnetic anisotropy (Fig. 1). TM clusters in the gas phase have also shown a strong dependence of the total magnetic moment ( $m_S + m_L$ ) on the particle size [3,25]. To date, however, fundamental points are still unclear. In particular, how does the magnetic anisotropy energy (MAE) evolve from single atoms to finite-sized magnetic particles, how does it correlate to the atomic magnetic moments, and how do both depend on the details of the atomic coordination. Such questions are

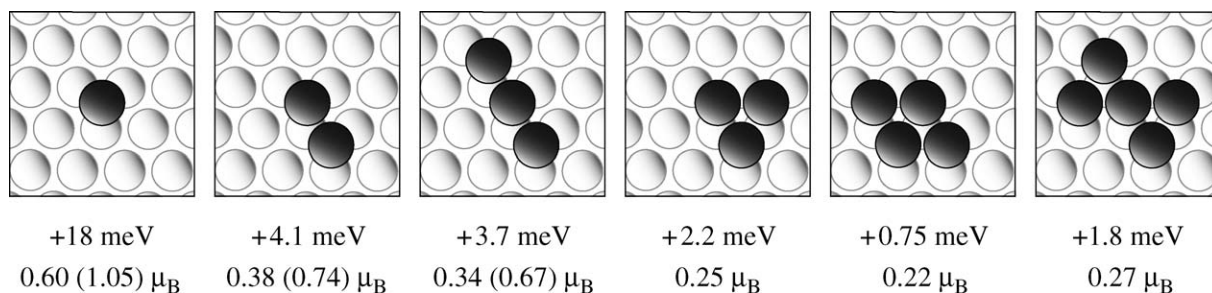


Fig. 1. Ab-initio calculation of  $E_a$  and  $m_L$  for Co on Pt(111) using the spin-polarized-relativistic Korringa–Kohn–Rostocker Green's function method in the spin-density approximation [30]. The Co atoms are on  $fcc$  sites. The values of  $m_L$  between brackets have been computed within the orbital polarization scheme with a 50% reduced Racah parameter. The reported values have been averaged over all Co sites for a given island, although site differences may well be relevant (see, e.g., [34]).

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