

# Lifetimes of magnetic excitations in Fe and Co atoms and clusters on Pt(111)

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Investigations of single magnetic atoms on a Pt surface revealed giant magnetic anisotropies. Recently, scanning tunneling microscopy was used to probe single Fe and Co atoms, dimers, and trimers on Pt(111). The magnetic anisotropy and, additionally, the lifetimes of the magnetically excited states were measured by inelastic tunneling spectroscopy. The lifetimes are in the order of femtoseconds due to an effective electron-electron relaxation process caused by the strong hybridization of the impurity states and the substrate. The different lifetimes are explained by the quantum mechanical nature of Fe and Co on Pt(111). The measurements of an Fe dimer show besides the collinear excitation, a noncollinear excitation with two possible decaying channels: spin-flip and non-spin-flip. Thus information on the magnetization dynamics can be extracted from inelastic spectra. © 2010 American Institute of Physics. [doi:10.1063/1.3365113]

## I. INTRODUCTION

In modern hard disks, the bits are stored in magnetically stable areas. Shrinking the bit area allows higher storage density. The smallest bit could ultimately be realized by a single magnetic atom. The stability of the bits is related to the energetic barrier to reverse their magnetization. In case of a single atom, the energy barrier is given by the magnetic anisotropy energy (MAE) of the atom. Gambardella *et al.*<sup>1</sup> found a giant MAE for Co atoms on Pt(111) of 9.3 meV with x-ray magnetic circular dichroism. However, this method cannot extract information about the magnetization dynamics, playing an equally important role for the stability of the bits. We use scanning tunneling microscopy (STM) with a high lateral resolution to probe single atoms, dimers, and trimers. The magnetization dynamics can be investigated with inelastic tunneling spectroscopy (ITS) and was also recently explored by theory.<sup>2</sup> In this report, the lifetimes of the excited magnetic states and the decay mechanisms are described.

## II. EXPERIMENT AND RESULTS

For preparation, small amounts of Fe or Co were deposited on an atomically clean Pt(111) crystal at 4.3 K, resulting in isolated atoms. The capabilities of atomic manipulation with the STM was used to create dimers and trimers.<sup>3</sup>

During the tunneling process, electrons may exchange angular momentum (spin) with the magnetic cluster or atom.<sup>4</sup> As a result of such an inelastic spin-flip scattering, the magnetization direction of the object is changed.<sup>5</sup> For Fe and Co atoms and clusters on Pt(111) the uniaxial anisotropy dominates, favoring a magnetization perpendicular to the surface plane.<sup>1,6,7</sup> The spin-flip of the tunneling electron

changes the spin of atom or cluster from the ground state  $S_z = \pm S$  to the excited state  $S_z = \pm (S-1)$ . The energy of the inelastic spin-flip process  $E_{sf}$  is the difference between these two states and can be measured with ITS. For metallic systems, elastic tunneling leads to a linear increase in tunneling current  $I$  upon increasing the bias voltage  $U$  in the low bias regime. When the energy  $eU$  reaches  $E_{sf}$ , an additional inelastic tunneling channel opens. As a consequence, the slope of  $I(U)$  is slightly increased for  $eU > E_{sf}$ . This change is usually too small to be identified directly. It can, however, be seen as a peak in  $d^2I/dU^2$ .<sup>8</sup> The excitation occurs for both tunneling directions such that a negative peak appears at negative bias.

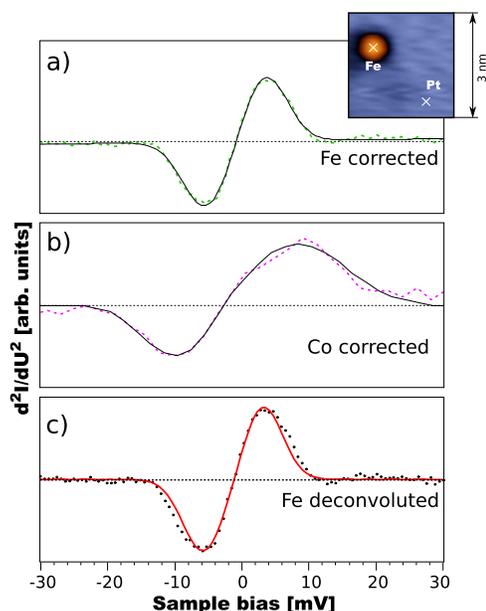


FIG. 1. (Color online) Topography of a single Fe atom on Pt(111) (inset), corrected  $d^2I/dU^2$  of Fe (a) and Co (b) atom and (c) raw Fe spectra (dots) with the deconvolution (solid red line).

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TABLE I. Spin relationship between measured excitation energies and MAE of Fe and Co atoms and clusters Ref. 7.

Cluster	Excitation energy (meV)	MAE (meV/atom)
Co <sub>1</sub>	10.25 ± 0.15	10.25 ± 0.15
Co <sub>2</sub>	8.2 ± 0.4	5.5 ± 0.3
Co <sub>3</sub>	8.3 ± 1.2	5.0 ± 0.8
Fe <sub>1</sub>	5.83 ± 0.08	6.53 ± 0.09
Fe <sub>2</sub>	5.98 ± 0.09	5.20 ± 0.09
Fe <sub>3</sub>	6.5 ± 0.2	5.4 ± 0.2

$d^2I/dU^2$  spectra were recorded for Fe atoms and the Pt surface (crosses in Fig. 1 inset). The genuine excitation spectrum (a) was obtained by subtracting the Pt background spectrum from the Fe spectrum. It is almost symmetric, with the minimum and the maximum reflecting inelastic magnetic excitations of an energy of 5.8 meV. For Co atoms on Pt(111), the same procedure provides  $E_{sf}$  of 10.3 meV [Fig. 1(b)].

The same experiments were done for Fe and Co dimers and trimers. Using the spin  $S=3/2$  and  $S=1$  for Fe and Co, respectively, and assuming a ferromagnetic coupling in the Fe and Co clusters, the classical MAE can be extracted from the observed excitation energies  $E_{sf}$  using the correspondence principle. All results are shown in Table I.

One notices that not only the position of the peaks changes for the Fe and Co spectra Fig. 1 [(a) and (b)], but also their widths. The measured signal  $f=d^2I/dU^2$  in the ITS experiment is a convolution of three functions: the intrinsic signal  $f_{in}$  is smeared with an experimental resolution function  $\phi$ , caused by the modulation voltage  $U_{mod}$  in the lock-in detection and the thermal resolution function  $\chi$  caused by the temperature  $T$ ,<sup>9</sup>

$$f = f_{in} \cdot \chi \cdot \phi.$$

Since both the temperature  $T$  and the modulation  $U_{mod}$  are known, the intrinsic signal  $f_{in}$  can be deconvoluted. In Fig. 1(c), the measured spectra (dots) and the deconvoluted (solid line) of an Fe atom is shown. They display the same peak position but different widths. The intrinsic width  $W_{in}$  is given by the lifetime  $\tau$  of the excitation via the uncertainty principle  $\tau W_{in} \gtrsim \hbar/2$ .  $W_{in}$  can be extracted from the deconvoluted spectra or directly from the measured width  $W$ ,

TABLE II. Lifetimes of excited states of atoms and clusters in femtoseconds.

	Atom	Dimer	Trimer	Dimer noncollinear
Fe	55	45	30	10
Co	20	45	30	

$$W_{in} = \sqrt{W^2 - (5.4kT)^2 - (1.7eU_{mod})^2}.$$

This procedure, applied to the observed excitations, gives the lifetimes as detailed in Table II. All the values are of the order of tens of femtoseconds. As the width of the peaks tends to disperse more than the peak position, the precision of the values is around  $\pm 5$  fs.

To relax the magnetically excited atom or cluster back to the ground state, spin angular momentum has to be transferred to the substrate. This can be achieved by spin-orbit interaction or much easier via spin-flip scattering of conduction electrons of the substrate, shown in Fig. 2(a) (dashed red arrows) for the case of Fe. This process is similar to the excitation mechanism. The electron that is scattered is, however, not provided by the tunneling current but by the Fermi sea of the substrate. Hence, an electron of the atom or cluster transfers  $\Delta S_z = \pm 1$  and energy to a conduction electron. The strong hybridization of the Fe or Co states with the Pt(111) substrate bands results in short lifetimes. The excitation lifetimes tend to decrease with increasing cluster size. The only exception from this rule is the lifetime of a single excited Co atom that is noticeably smaller than the value for the Fe atom. The reason for this effect lays in the spin configuration of the excited state [Fig. 2(b)]. An excited Co atom has  $S_z = 0$  and can thus relax into both ground states with  $S_z = \pm 1$ . All the other clusters have a spin higher than 1 and the excited state can only relax in one direction. This would effectively halve the lifetime of the excited state of Co with respect to other clusters, in good agreement with the measured values. A further reduction of the lifetime is caused by a stronger hybridization of Co with the substrate, as manifested by the higher MAE values.

In the case of an Fe dimer, the total dimer spin is composed of the two spins of the individual atoms. In the ground state, both spins align in parallel. For simplicity, we assume that both spins have a maximal spin of  $s=3/2$ . The interaction with an electron leads to a transition from a ground state

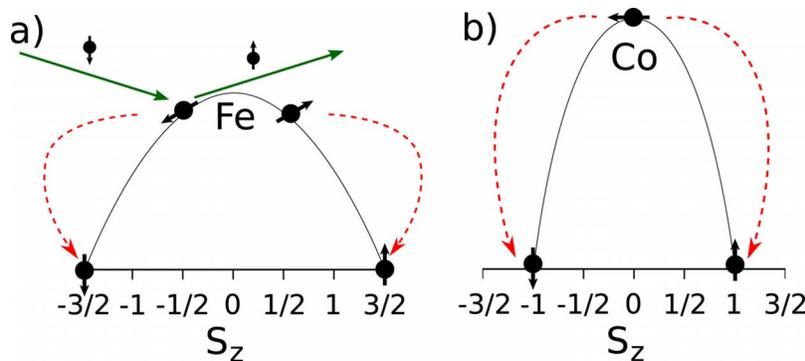


FIG. 2. (Color online) Scheme of relaxation processes for (a) Co and (b) Fe atoms.

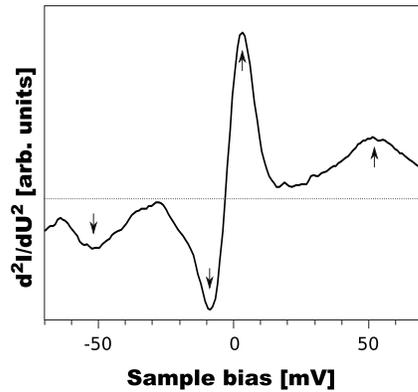


FIG. 3. Inelastic spectrum taken on an Fe dimer, with two inelastic excitations visible at  $\pm \approx 5$  meV and  $\pm \approx 50$  meV as indicated by arrows.

$|s, s\rangle$  to a superposition of states, where the spin of one of the composing atoms is changed by one, namely  $|s, s-1\rangle$  and  $|s-1, s\rangle$ . There are two linear combinations of these states that form an eigenstate of the Heisenberg Hamiltonian including the exchange constant  $J$ ,

$$\Psi_1 = \frac{1}{\sqrt{2}}|s, s-1\rangle + \frac{1}{\sqrt{2}}|s-1, s\rangle, \quad (1)$$

$$\Psi_2 = \frac{1}{\sqrt{2}}|s, s-1\rangle - \frac{1}{\sqrt{2}}|s-1, s\rangle. \quad (2)$$

The energies of the two states differ by  $3J$ , so two inelastic excitation signatures are expected in the spectrum.

The cluster in the first state has a total spin of  $S=2s$  and the  $z$  projection  $S_z=2s-1$ . Thus, this state can be described as a rotation of the total spin of the cluster in the analogy to the excitation of an individual atom. The energy of this excitation can therefore be related to the classical MAE of the whole cluster as described before. Classically, this state is an in-phase precession of the two atomic spins.

In the second state, the dimer has a spin of  $S=S_z=2s-1$ . Viewed classically, the time evolution of this state is equivalent to an out-of-phase precession of the two spins. The energy of the state is higher due to the exchange interaction between the two spins coming into play. The total energy cost of this excitation is a sum of the exchange and the anisotropy contributions,  $E_x=2Js-D(4s-1)$ . As the exchange constant  $J$  is of the order of tens of  $\text{meV}/\hbar^2$  in bulk Fe,<sup>10</sup> one can expect an inelastic peak around 100 meV. And indeed such a peak is observed, as shown on Fig. 3. An excitation energy of  $54 \pm 2$  meV is observed, giving a  $J$  of  $16 \pm 1$   $\text{meV}/\hbar^2$ . The noncollinear state has the shortest lifetime of all. A likely reason for this is given by two possible ways for decaying: spin-flip and non-spin-flip processes. The spin-flip process is the inverse process of the excitation similar to the relaxation discussed above, relaxing the dimer to the ground state (see dashed red arrows Fig. 4). Due to conservation of spin moment, only the majority electrons can relax the cluster. The non-spin-flip relaxation leads to a decay to the collinear excited state, keeping  $S_z$  but changing the phase of the dimer spin wave function. Thus electrons of both spins can de-excite the noncollinear dimer state via non-spin-flip scattering, doubling the scattering probability. Note

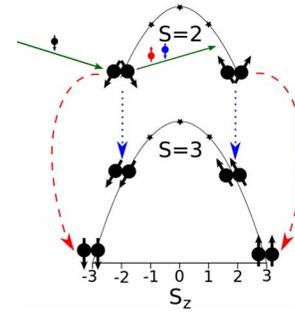


FIG. 4. (Color online) Scheme of relaxation processes for an Fe dimer.

that the substrate is nonmagnetic, i.e., the Pt bands are spin degenerate. As a consequence, the available phase space for spin-flip and non-spin-flip scattering is identical. The different relaxation mechanisms are illustrated in Fig. 4, dashed red arrows indicate spin-flip scattering while dotted blue arrows indicate non-spin-flip scattering. The second process is more probable, as discussed above, in agreement with the shorter lifetimes of the noncollinear excited state.

### III. CONCLUSION

These lifetimes give information on the magnetization dynamics of the atoms and clusters. The extremely short lifetimes in these metallic systems indicate an efficient relaxation process, which is absent for magnetic ions in insulator structures.<sup>11</sup> Since comparable MAEs were found in our metallic structures and in insulator structures, one can exclude the responsibility of the spin-orbit interaction for the short lifetimes in the metallic case. More likely, the strong hybridization of the atomic states with the Pt substrate states leads to efficient electron-electron scattering processes that relax the magnetic state of the atom.

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<sup>1</sup>P. Gambardella, S. Rusponi, M. Veronese, S. S. Dhesi, C. Grazioli, A. Dallmeyer, I. Cabria, R. Zeller, P. H. Dederichs, K. Kern, C. Carbone, and H. Brune, *Science* **300**, 1130 (2003).

<sup>2</sup>J. Fransson, *Nano Lett.* **9**, 2414 (2009).

<sup>3</sup>D. M. Eigler and E. K. Schweizer, *Nature (London)* **344**, 524 (1990).

<sup>4</sup>A. J. Heinrich, J. A. Gupta, C. P. Lutz, and D. M. Eigler, *Science* **306**, 466 (2004).

<sup>5</sup>T. Balashov, A. F. Takács, W. Wulfhekkel, and J. Kirschner, *Phys. Rev. Lett.* **97**, 187201 (2006).

<sup>6</sup>C. Etz, J. Zabloudil, P. Weinberger, and E. Y. Vedmedenko, *Phys. Rev. B* **77**, 184425 (2008).

<sup>7</sup>T. Balashov, A. F. Takács, A. Ernst, S. Ostanin, J. Henk, I. Mertig, P. Bruno, T. Miyamachi, S. Suga, and W. Wulfhekkel, *Phys. Rev. Lett.* **102**, 257203 (2009).

<sup>8</sup>B. C. Stipe, M. A. Razaeei, and W. Ho, *Science* **280**, 1732 (1998).

<sup>9</sup>J. Klein, A. Léger, M. Belin, D. Défourneau, and M. J. L. Sangster, *Phys. Rev. B* **7**, 2336 (1973).

<sup>10</sup>M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, *Phys. Rev. B* **64**, 174402 (2001).

<sup>11</sup>C. F. Hirjibehedin, C.-Y. Lin, A. F. Otte, M. Ternes, C. P. Lutz, B. A. Jones, and A. J. Heinrich, *Science* **317**, 1199 (2007).