Confused Hund's Rules in the Induced Magnetic Moments in 5d Layers interfaced with bcc Fe(001)

Recent technological developments in magnetic recording and data storage have increased interest in studying electronic structure and magnetic properties of thin films, multilayers and nanostructures, both theoretically and experimentally. Advances in synchrotron radiation instrumentation have made it possible to determine the electronic and magnetic properties of these systems in an element specific way. X-ray circular magnetic dichroism (XMCD), in conjunction with the sum rules, enables the separation of spin and orbital contributions to the total magnetic moments. In addition, complementary soft-X-ray resonant magnetic scattering measurements can provide details about the layer dependence of the magnetic moments, periodicity of the magnetic domain structures, and roughness of the magnetic layers. The present day relativistic ab initio theory can not only assist experiments, but is able to study trends and systematics as well as make important predictions, which is demonstrated in the present highlight.

Recently, Wilhelm et al. [1] reported XMCD measurements at the W and Ir $L_{2,3}$ edges of W/Fe and Ir/Fe multilayers. The 5$d$ spin and orbital magnetic moments, $\mu_S$ and $\mu_L$, of W and Ir were deduced by applying the sum rules. These moments are induced in the 5$d$ band, through hybridization, by the spin polarization of the Fe, which is reflected in their small values. For Ir, $\mu_S$ and $\mu_L$ align both parallel to the Fe spin moment, while for W they align both anti-parallel to Fe. As a result, it was concluded that in the case of W, with its less than half-filled 5$d$ band, this implies a violation of the Hund's third rule. In addition, the authors concluded that the induced magnetic behavior of 5$d$ layers might be radically different than that of impurities and alloys. Here we show, on the basis of a systematic set of ab-initio calculations, that the behavior of $\mu_S$ and $\mu_L$ of 5$d$ magnetic impurities in an Fe host is actually very similar to that of 5$d$ interface layers in Fe multilayers, proving that the conclusion, inferred on the basis of two data points, is incorrect.

Calculations of the electronic and magnetic properties were performed for periodic multilayers, Fe/Z, consisting of 5 monolayers (ML) of Fe(100) and 3 ML of the 5$d$ transition metal, Z, with Z spanning the whole 5$d$ series [2], using the relativistic ab-initio spin-polarized linear muffin-tin orbitals method [3]. Figure 1 includes, besides the presently calculated values of spin and orbital moments for the 5$d$ interface layers [2], also the results of two different impurity calculations [4, 5].

The Hund's third rule states that spin and orbital moments should be parallel (antiparallel) for more (less) half-filled shell. Although strictly valid only for single atoms, this rule seems to be also applicable to solids, however with a few exceptions. In Fig. 1, it is seen that the spin moment $\mu_S$ of 5$d$ impurities in Fe [4, 5] is aligned anti-parallel for the first part of the 5$d$ transition metal series, i.e. for less than half filled band. From Os onwards $\mu_S$ of the 5$d$ impurity and Fe are parallel aligned. For 5$d$ impurities to obey Hund's third rule, the orbital moment $\mu_L$ should be positive throughout the 5$d$ series: this is not the case for Re, Os, and Ir. The curves of the calculated $\mu_S$ and $\mu_L$ for the multilayers show a similar behavior as for the impurities. The orbital moment $\mu_L$, being more sensitive to the structure and chemical environment, switches between parallel and anti-parallel at nearly the same location, resulting in a negative value for W, Re, and Os. Therefore, the relative alignment of
\( \mu_S \) and \( \mu_L \) in the 5\textit{d} metals is the same as in the single impurity case, with the exception of W and Ir.

The present calculations agree well with the experimental data which are for \( \mu_S \) and \( \mu_L \) (\(-0.18; -0.016\)) and (0.20; 0.019) in W and Ir respectively, while the theory gives (\(-0.11; -0.031\)) and (0.26; 0.003). It just so happens that in the vicinity of W and Ir a sign change of the orbital moment \( \mu_L \) occurs (Fig.1). This shows that conclusions regarding systematics should have not been drawn on the basis of these two data points alone. Our results indicate that mostly band filling, and to a lesser extent geometrical effects, determine the trend of \( \mu_S \) and \( \mu_L \) over the 5\textit{d} series. This finding is corroborated by the fact that a similar trend also occurs for the 4\textit{d} series. [2]

FIG. 1: Calculated spin magnetic moments (\( \mu_S \), top panel) and orbital magnetic moments (\( \mu_L \), lower panel) in \( \mu_B/\text{atom} \) for 5\textit{d} interface layers in 5Fe(100)/3Z multilayers, including interface relaxations, (Ref. [2]: ⋄) compared to 5\textit{d} impurities in Fe (Ref. [4]: □ and Ref. [5]: Δ)