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Ji-Hyun Kim¹, avel Lukashev², Serl-Un Wang¹, Jae-Sung Kim¹*, Xumin Chen³, Geoffrey Rojas³, Violetta Sessi⁴, Jan Honolka⁵, Ralph Skomski³, Renat F Sabirianov², and Axel Enders³

¹Department of Physics, Sookmyung Women's University, Seoul, 140-741, South Korea

²Department of Physics, University of Nebraska at Omaha, Omaha, NE 68182

³Department of Physics and Astronomy, University of Nebraska-Lincoln, Lincoln, NE 68588

⁴European Synchrotron Radiation Facility, 6 Rue Jules Horowitz, BP 220, 38043 Grenoble Cedex 9, France

⁵Max Planck Institute for Solid State Physics, Heisenbergstr. 1, Stuttgart, 70569, Germany

Magnetic nanostructures often exhibit spin configurations that do not occur in nature. For example, iron is a prototypical ferromagnet in its bulk *bcc* phase but exhibits low-spin—high-spin transitions, antiferromagnetism, complex spin structures and even noncollinear magnetic order in thin films and other nanostructures [1, 2, 3, 4, 5]. This diversity is especially pronounced for dense-packed fcc-likeatomicenvironments [6]. By contrast, metallic cobalt tends to exhibit stable ferromagnetism, even in dense-packed atomic arrangements [7] and in nanostructures [8]. Here we show that small clusters of Co, formed on a crystalline W (110) surface and containing 3 to 12 atoms, are ferri-, ferro- or nonmagnetic, depending on cluster size and geometry. First-principle calculations are used to determine the atomic Co moments and their relative alignment, and showed that antiferromagnetic spin alignment in the Co clusters is due to a combined effect of hybridization with the tungsten substrate and band filling. Our theoretical predictions are supported by X-ray magnetic circular dichroism measurements of such Co clusters [Fig. 1]. The Co clusters, which are synthesized on reconstructed W (110) template surfaces [Fig. 2], exhibit an average magnetization, which is much smaller than expected for ferromagnetic Co moments and consistent with the proposed theoretical predictions. By performing a comparative parallel study on the isostructural Fe/W(110), we show that the effect occurs for Co but not for Fe.

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Fig. 1 XAS spectra (top) and XMCD spectra (bottom) taken at the L3/2 adsorption edge of Fe and Co (T = 8 K, B = 5.5 T). The measurements on the right were done on 0.02 ML Co/W (110), and those on the left after 0.2 ML Fe had been added to this sample. The reference measurement, shown as blue line, was taken on 0.05 ML Co on Pt (111) at 10 K. (adapted from ref. [9]).



Figure 3. (a) STM images of the reconstructed, carbureated W (110); (b) Co clusters on C/W (110); (c) The spacing between adjacent Co cluster rows can be controlled via the W (110) reconstruction. (d) Deposition of Fe to pre-existing Co clusters results in Fe-Co core-shell islands arranged in rows along the <111> direction, marked by red arrows, and in pristine Fe clusters such as those circled in blue.