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Control of Magnetic Order in Monolayer Films by Substrate Tuning STEFAN HEINZE, PAOLO FERRIANI, Institute of Applied Physics, University of Hamburg, Germany, ILJA TUREK, Department of Electronic Structures, Charles University, Prague, Czech Republic, GUSTAV BIHLMAYER, STEFAN BLÜGEL, Institut fuer Festkoerperforschung, Forschungszentrum Juelich, Germany — Surprisingly, antiferromagnetic order has recently been observed in a monolayer (ML) film of Fe on W(001) [1] and a novel, nanoscale magnetic structure has been discovered for a ML Fe on Ir(111) [2] showing the crucial influence of the substrate. Here, we therefore propose to tailor exchange interactions in magnetic monolayer films by tuning the adjacent non-magnetic substrate. Using first-principles calculations based on density functional theory, we demonstrate a ferromagnetic-antiferromagnetic phase transition for one ML Fe on a Ta_x $W_{1-x}(001)$ surface as a function of the Ta concentration. At the Ta concentration of the transition, the nearest-neighbor exchange interaction becomes negligible and exchange terms beyond nearest-neighbors and higher order spin interactions beyond the Heisenberg Hamiltonian become crucial. In this regime, the accessible magnetic phase space is dramatically enhanced, and we study complex magnetic order such as spin-spiral states, multiple-q states, or even disordered local moment states. [1] A. Kubetzka, *et al.*, Phys. Rev. Lett.**94**, 087204 (2005). [2] K. von Bergmann, *et al.*, Phys. Rev.Lett. **96**, 167203 (2006).

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