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Emergence and Light-Driven Tuning of a Correlated Ferromagnetic Glass in FM/Molecule Hybrid Systems

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Thin *3d* ferromagnetic metals (FM) thin films interfaced with organic molecules can exhibit strong modification of their magnetic properties, like anisotropy and magnetic moment [1].These effects are understood as ultimately caused by the surface FM *d* orbitals hybridized with the molecular *p* orbitals, altering their contribution to wavefunction-related quantities like the Spin-Orbit Coupling [2,3].

In my talk I will discuss theoretical and experimental evidences that the effects induced by the interfacial hybridization are not only quantitative but also cause the collapse of the standard magnetic domain structure and the establishment of a Correlated Ferromagnetic Glass (CGS) [4]. Few-nm-thick Co films are interfaced with a wide variety of molecular (MOL) layers, including fullerene (C_{60}), Gallium-quinoline (Gaq_3) and sexithiophene (T6), and characterized by MOKE, AMR, SQUID and MFM measurements. A significant magnetic hardening is observed[4,5], detectable even at room temperature. Additionally, minor loops characterization of Co/Mol samples reveals an unusual low-field magnetization dynamics, deviating from the standard domain nucleation and motion, derived from the Rayleigh law[6,7]. These results are explained by a joint DFT-micromagnetic model, showing that the adsorbed molecules induce an additional local in-plane uniaxial anisotropy, randomly oriented but correlated over distances (r_c) comparable to domain wall widths (Δ). The magnetic configuration turns from a domain-like to a glassy-like type, where no distinction can be made between domain and domain walls. Such configuration presents a number of localized out-of-plane vortices, successfully detected by high-resolution MFM imaging.

I will also show that the interface-driven magnetic properties can be actively controlled by optical means, using Co/C_{60} heterostructures as a prototypical system [8]. By creating excitons in the molecular layer with resonant ultrashort light pulses, we achieve a 60% reduction of the magnetization precession frequency, ultimately due to a quenching of the interface-induced magnetic anisotropy.

[1] M. Cinchetti et al. Nat. Mater. **16** (2017) 507.
[2] K. Bairagi et al., Phys. Rev. B **98** (2018) 085.
[3] A. Halder et al., Phys. Rev. Mat. **7** (2022) 064409.
[4] M. Benini et al, arXiv:2503.15749 (2025).
[5] M. Benini et al., Adv. Mater. Interfaces **9** (2022) 2201394.
[6] Takahashi S et al., Philosophical Magazine **91** (2011) 2216.
[7] Rayleigh L., LED Phil. Mag **23** (1887) 225.
[8] M. Benini et al, arXiv:2504.00551 (2025).