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Controlled Under Pressure: Understanding Spin Orbit Coupling and Exchange Anisotropy in Organic Magnets[1]

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The application of high pressure in the study of molecule-based materials has gained considerable interest, in part due to their high compressibilities, but also because the relevant electronic/magnetic degrees of freedom are often very sensitive to pressure. For example, small changes in the coordination environment around a magnetic transition metal ion can produce quite dramatic variations in both the on-site spin-orbit anisotropy as well as the exchange interactions between such ions when assembled into clusters or 3D networks. This has spurred the development of sophisticated spectroscopic tools that can be integrated with high-pressure instrumentation. The study of magnetic structure/property relations requires not only precise crystallographic data, but also detailed spectroscopic information concerning the unpaired electrons that give rise to the magnetic properties. This invited talk will begin with a brief description of the development and application of methods enabling EPR studies of oriented single-crystal samples subjected to hydrostatic pressures of up to 3.5 GPa. After an introductory example,[3] the remainder of the talk will focus on a family of heavy atom organic radical ferromagnets (containing S and Se heteroatoms) that hold records for both the highest transition temperature and coercivity (for organic magnets). The latter is the result of an unexpectedly high magnetic anisotropy, attributable to spin-orbit-mediated exchange (hopping) processes.[4,5] Ferromagnetic resonance (FMR) measurements reveal a continuous increase in the magnetic anisotropy with increasing pressure in the all Se compound, in excellent agreement with ab initio calculations based on the known pressure-dependence of its structure.[6] The large value of anisotropic exchange terms in this heavy atom organic ferromagnet emphasizes the important role of spin-orbit coupling in a wide range of organics where this effect is usually considered to be small.

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[3]Prescimone et al., Angew. Chem. Int. Ed. **51**, 7490 (2012).

[4]Winter et al., J. Am. Chem. Soc. **133**, 8126 (2011).

[5]Winter et al., Phys. Rev. B **85**, 094430 (2012).

[6]Thirunavukkuarasu et al., submitted (2014).