

Controlling Anisotropy in Organic-Based Magnets for Microwave Electronics and Quantum Magnonics

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Progress in the field of organic electronics has yielded significant advances in the development and application of organic light emitting diodes (OLEDs), organic photovoltaics (OPVs), and organic field effect transistors (OFETs). The success of these device applications suggests that further expansion of the field to include magnetic functionality may be promising. At the same time, the emergence of optimized thin film growth of and successful encapsulation strategies for organic magnetic materials allows for long term stability of high quality magnets under ambient conditions. Organic-based magnets of the form $M[\text{Acceptor}]_x$ ($M = \text{transition metal}$, $x \approx 2$) exhibit room temperature magnetic ordering and provide the opportunity to tailor magnetic properties through the careful selection of the transition metal ions and organic ligands. However, despite the promise of these systems, important outstanding questions remain. For example, while these materials are typically considered to be globally disordered structurally, the canonical magnet of this materials class, vanadium tetracyanoethylene ($V[\text{TCNE}]_x$; $x \approx 2$), exhibits robust magnetism, has a single-peaked, ultra-narrow magnetic resonance feature (approximately 1 G at 10 GHz), and has been shown to have a Curie temperature as high as 600 K.

Here, we present work that illustrates strategies to control the anisotropy in this material system through growth morphology or temperature-dependent structural changes. We present the facile synthesis of a new class of organic-based magnetic nanostructures consisting of nanowires of $V[\text{TCNE}]_x$ that self-assemble along the ridges of a grooved substrate. These nanowires exhibit uniaxial magnetic anisotropy in direct contrast to the isotropic in-plane response of typical thin-films. Furthermore, the magnetic anisotropy persists to the point of re-coalescence of the nanowires into a thin film, suggesting that dipole fields arising from shape anisotropy do not play a dominant role and instead pointing to interface-templated structural ordering as the origin of this anisotropy. Moreover, isolated films of the $V[\text{TCNE}]_x$ analogs vanadium methyl tricyanoethylenecarboxylate ($V[\text{MeTCEC}]_x$) and vanadium ethyl tricyanoethylene carboxylate ($V[\text{ETCEC}]_x$) show a temperature-dependent switch in the easy axis from in-plane to out of plane, again suggesting underlying effects beyond shape anisotropy and pointing towards the presence of an additional contribution to the uniaxial anisotropy. These results introduce a new degree of freedom for organic-based magnetism and spintronics, allowing for the engineering of magnetic anisotropy in a material that exhibits both robust room-temperature magnetic order and the benefits of low cost, mechanical flexibility, and facile synthesis found in other organic materials. Together with recent demonstration of encapsulation technologies that allow for device operation under ambient conditions and demonstrated functional microwave devices that exhibit high quality factors across a frequency range of 1–20 GHz, these results suggest future promising applications in microwave electronics and quantum magnonics.