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# Conducting molecular nanomagnet of Dy(III) with partially charged TCNQ radicals

Xuan Zhang,<sup>[a]</sup> Haomiao Xie,<sup>[a]</sup> Maria Ballesteros-Rivas,<sup>[a]</sup> Toby J. Woods,<sup>[a]</sup> and Kim R. Dunbar\*<sup>[a]</sup>

**Abstract:** Bifunctional electrically conducting single-molecule magnets are highly promising platforms for non-volatile memory devices and quantum computing applications. The development of these molecular materials, however, has largely been hindered by the lack of straightforward synthetic methods. Herein we demonstrate a facile and modular approach for the realization of bifunctional materials that does not require electrochemical or chemical oxidation to obtain partially charged organic radicals. Magnetic and electrical conductivity studies reveal that the Dy(III) compound exhibits slow relaxation of the magnetization between 5.0-8.0 K and semiconducting behavior over the range 180-350 K. DC magnetic fields suppress the quantum tunneling of the magnetization and affect the spin-canted antiferromagnetic interactions.

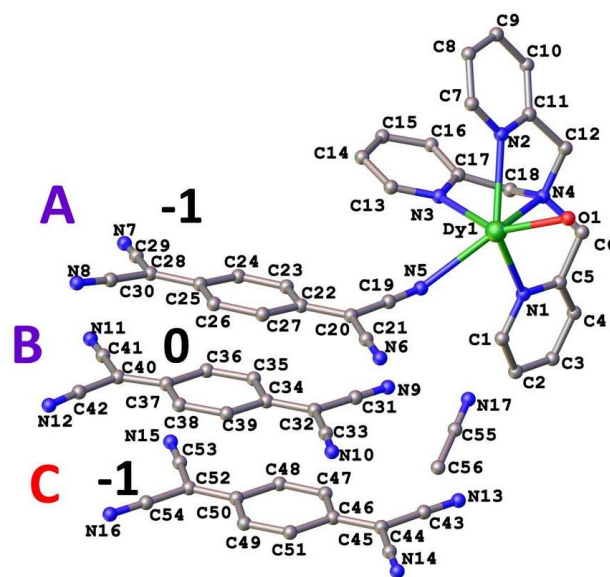
In the quest for an alternative to conventional electronic materials, which are estimated to approach their physical miniaturization limit by the early 2020s, the design and synthesis of molecular nanomaterials for electronic devices have been gathering increasing attention over the past two decades.<sup>[1]</sup> One of the most prominent candidates are molecular nanomagnets that are capable of exhibiting remnant magnetization much like a traditional bulk magnet, but with a much smaller size such as 1 or 2 nanometers.<sup>[1c, 1d, 2]</sup> As a result, such molecular nanomagnets can provide higher density information storage, operate at much faster speeds and require less energy than conventional devices because of the quantum effects that they exhibit.<sup>[3]</sup>

The application of nanomaterials to the elaboration of new types of electronic devices presents numerous challenges, one of which is the synthesis of bifunctional molecular materials that exhibit both single-molecule magnet (SMM) and semiconducting properties. By constructing such materials with alternating functional layers, one can address their magnetic behavior through the application of an electric field, or vice versa, which will make them superior candidates for transistors and sensors. By using two distinct inorganic and organic components, we and others have achieved crystalline electrical conducting spin-crossover (SCO) materials.<sup>[4]</sup> For instance, we observed synergistic interactions in a bifunctional SCO electrical conductor composed of alternating layers of  $[\text{Co}(\text{terpy})_2]^{2+}$  spin-crossover species and partially charged TCNQ radicals.<sup>[4a]</sup> The realization of this material is an important step towards potential device applications as they can be processed directly from solution and crystallize in only a few minutes.

Reports on bifunctional electrical conducting SMMs are scarce, owing largely to the lack of simple and stable SMM candidates and readily accessible partially charged radical source and hence a facile modular synthetic approach that combines a cationic

SMM species with an anionic electrically conducting sub-lattice. Previously reported synthetic strategies for realizing significant electrical conductivity are non-trivial as they require electrochemical oxidation to obtain partially charged metal-organic radicals.<sup>[5]</sup> In this vein, our group reported the incorporation of TCNQF<sup>-</sup> radicals in a [Tb-Cu] molecular nanomagnet system, but due to the localization of electron density on TCNQF<sup>-</sup>, no significant electrical conductivity was observed.<sup>[6]</sup>

In considering targets for the incorporation of paramagnetic molecules into multifunctional materials we note that Dy(III) compounds have been widely studied as candidates for SMMs due to their high magnetic anisotropy. In fact, the record for blocking temperatures and effective energy barriers are held by mononuclear Dy(III) compounds<sup>[7]</sup>; SMMs composed of dinuclear Dy<sub>2</sub> units are also quite interesting as candidates for the realization of high effective energy barriers owing to ferromagnetic magnetic interactions between the Dy(III) ions.<sup>[3a, 8]</sup>



**Figure 1.** Asymmetric unit of  $[\text{Dy}(\text{TPMA})(\mu\text{-TCNQ})(\mu\text{-OH})](\text{TCNQ})_2 \cdot \text{CH}_3\text{CN}$  (1). Hydrogen atoms are omitted for the sake of clarity.

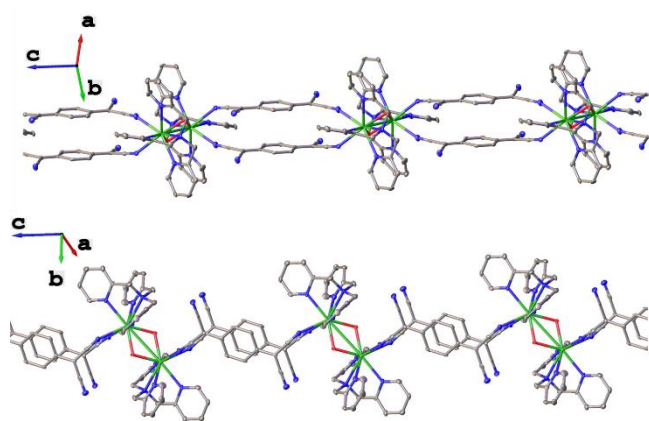
Herein we report the convenient use of the partially charged TCNQ radical source  $(\text{Et}_3\text{NH})(\text{TCNQ})_2$  as a precursor for a series of isostructural chains bridged by hydroxide groups and TCNQ radicals,  $\{[\text{Ln}(\text{TPMA})(\mu\text{-OH})(\mu\text{-TCNQ})](\text{TCNQ})_2 \cdot \text{CH}_3\text{CN}\}_n$  (Ln = Dy (1), Gd (2) and Y(3); TPMA = tris(2-pyridylmethyl)amine). Magnetic and electrical conductivity studies reveal that the Dy(III) compound exhibits slow relaxation of the magnetization between  $T = 5\text{-}8$  K and semiconductivity from  $T = 180\text{-}350$  K. As far as we are aware, this is the first TCNQ radical-based electrical conducting material to exhibit slow relaxation of the magnetization reported to date.

[a] Dr. X. Zhang, H. Xie, Dr. M. Ballesteros-Rivas, Dr. T. Woods, Dr. K. Dunbar  
Department of Chemistry  
Texas A&M University  
P.O. Box 30012, College Station, TX 77842-3012 (USA)  
E-mail: dunbar@chem.tamu.edu

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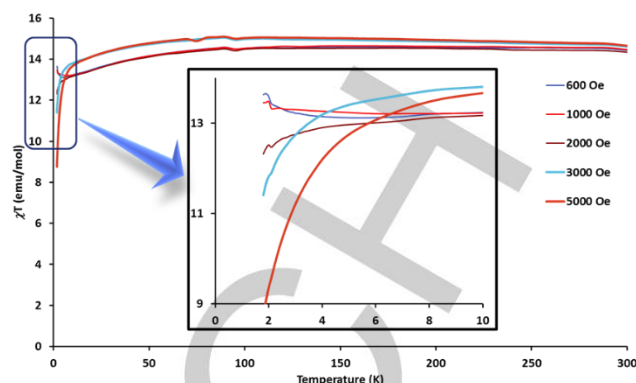
## COMMUNICATION

Dark purple crystals of **1** form in the time span of minutes when a solution of  $(\text{Et}_3\text{NH})(\text{TCNQ})_2$  in acetonitrile was added to a solution of Dy(III) trifluoromethanesulfonate and TPMA in acetonitrile. The product crystallizes in the triclinic space group  $P\bar{1}$  (ESI, Table S1) with the asymmetric unit being composed of a Dy(III) ion coordinated to the TPMA ligand, one TCNQ radical and one hydroxide anion; in addition, there is one uncoordinated TCNQ species and an acetonitrile molecule (Figure 1). The Dy(III) ion is eight coordinate with four nitrogen donors from the TPMA ligand, two nitrogen atoms from two bridging TCNQ species and two oxygen atoms from the two bridging hydroxide groups. The Dy(III) ions are bridged by hydroxide and TCNQ species to form 1-D chains of  $\{[\text{Dy}(\text{TPMA})(\mu\text{-TCNQ})(\mu\text{-OH})_2]_2\}_\infty$  along the  $c$  axis (Figure 2). The bond angle of Dy-O-Dy is  $111.5(2)^\circ$  and the distance between the two Dy(III) ions that are bridged by the hydroxide group is  $3.6901(9)$  Å. The two  $\mu_2$ -TCNQ anions exhibit a short stacking distance of  $3.273(2)$  Å. The  $\{[\text{Dy}(\text{TPMA})(\mu\text{-TCNQ})(\mu\text{-OH})_2]_2\}_\infty$  chains are then organized via  $\pi$ - $\pi$  stacking interactions of the TCNQ species (Figure S3). The TCNQ species have an average charge of  $-2/3$ , as two of them are monoanions and the third one is neutral, as estimated from Kistenmacher's empirical formula (Figure 1 and Table S2).<sup>[9]</sup>



**Figure 2.** Perspective views of the structure of **1** showing the bridging TCNQ radicals (top) and the bridging hydroxide groups (bottom) in the  $\{[\text{Dy}(\text{TPMA})(\mu\text{-TCNQ})(\mu\text{-OH})_2]_2\}_\infty$  chains.

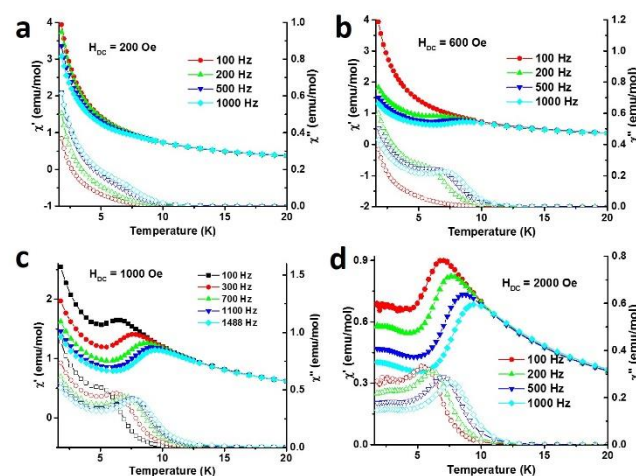
Temperature dependent magnetic susceptibility measurements of **1** were carried out under a DC field of 1000 Oe over the temperature range of 300–1.8 K (Figure 3 and Figure S4). The  $\chi T$  value remains nearly constant at  $14.6 \text{ emu}\cdot\text{K}\cdot\text{mol}^{-1}$  (per formula unit), until  $\sim 100$  K, in accord with the expected contributions from one Dy(III) ion and one TCNQ $^-$  (Dy(III),  $4f^9$ ,  $^6\text{H}_{15/2}$ ,  $J = 15/2$ ,  $L = 5$ ,  $S = 5/2$ ,  $g = 1.33$ ; TCNQ $^-$ ,  $S = 1/2$ ,  $g = 2.0$ ). Below 100 K, the  $\chi T$  value gradually decreases to a minimum value of  $13.2 \text{ emu}\cdot\text{K}\cdot\text{mol}^{-1}$  at  $\sim 6$  K which can be due to antiferromagnetic coupling between the spin centers and/or the thermal depopulation of the excited  $M_J$  states of Dy(III) ions as the temperature decreases. Upon further lowering the temperature, the  $\chi T$  value exhibits an increase to a maximum of  $13.5 \text{ emu}\cdot\text{K}\cdot\text{mol}^{-1}$  at 2 K, indicating the presence of weak



ferromagnetic interactions between the spin centers (*vide infra*). Below 2 K, the  $\chi T$  value drops slightly to  $13.4 \text{ emu}\cdot\text{K}\cdot\text{mol}^{-1}$ .

**Figure 3.** The temperature dependence of the  $\chi T$  product of **1** under different applied DC magnetic fields. The inset shows the low temperature region.

The expected magnetic moment for one Dy(III) and one TCNQ $^-$  radical is  $\sim 12.4$  Bohr magnetons but the observed magnetic moment of **1** is  $\sim 5.8$  Bohr magnetons at 1.8 K and 7 T (Figure S5). The lack of magnetization saturation even at 7 T in the field-dependent magnetization data at 1.8 K supports the fact that there is significant magnetic anisotropy for the compound.



**Figure 4.** Temperature dependence of the in-phase (solid symbols) and out-of-phase AC magnetic susceptibilities of **1** under applied DC magnetic fields of 200 (a), 600 (b), 1000 (c) and 2000 (d) Oe.

AC magnetic susceptibility measurements were performed to probe the dynamic magnetic properties of **1**. Under a zero applied DC field, only the beginning of an out-of-phase ( $\chi''$ ) AC signal is observed down to 1.8 K (Figure S6), which is attributed to quantum tunneling.<sup>[10]</sup> When DC magnetic fields of different magnitudes were applied for the AC measurements, the degeneracy of the  $\pm M_J$  states are lifted and a series of frequency-dependent peaks in the out-of-phase ( $\chi''$ ) signals appear at 5–8 K (Figure 4). At the optimized DC field of 2000 Oe, quantum tunneling of the magnetization (QTM) is minimized and a slow

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relaxation process is observed (Figure 4d and Figure S7) which is solely Orbach in nature as evidenced by the fact that the relaxation time can be fit to an Arrhenius law to yield an effective energy barrier for the reversal of magnetization of 43.4 K with a pre-exponential factor of  $\tau_0 = 7.8 \times 10^{-7}$  s (Figure S8). This thermal relaxation process is attributed to the magnetic anisotropy of the  $[\text{Dy}_2(\mu\text{-OH})_2]^{4+}$  units in the compound.<sup>[11]</sup> The faster relaxation process at lower temperatures are consistent with inter-chain interactions, which is still operative in the  $T = 5\text{-}8$  K region at 2000 Oe. Therefore, two modified Debye functions were employed to fit the experimental Cole-Cole plot over this temperature range (Figure S9) using CC-FIT.<sup>[12]</sup> The dominant thermal relaxation process exhibits  $\alpha$  values between 0.12 and 0.06, indicating a narrow distribution of the relaxation times (Table S5).

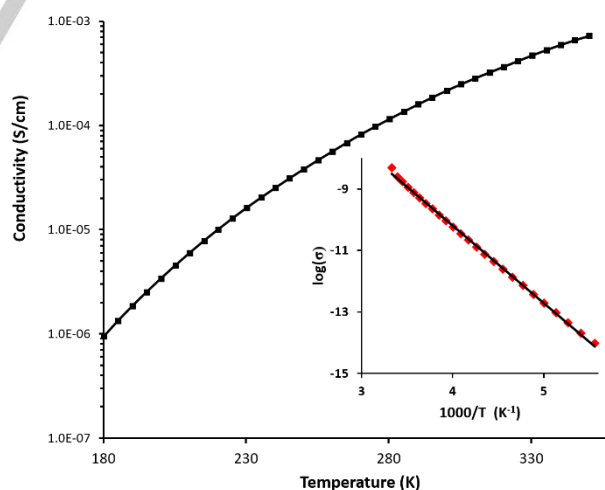
Given that the dynamic magnetic properties of **1** exhibit dependence on the DC magnetic field, its temperature dependent magnetic susceptibilities were also investigated under various DC magnetic fields in order to probe the effect of external DC fields on the magnetic interactions in **1**. Below 2000 Oe, the  $\chi T$  values display pronounced increases at  $\sim 2$  K, indicating the presence of weak ferromagnetic interactions at low fields (Figure 3). Another feature that is worth noting is that the maxima in the  $\chi T$  plots at 2 K diminish as the fields are increased and disappear when the fields are greater than 3000 Oe, indicating that the weak ferromagnetic interactions are suppressed by the application of external DC magnetic fields. These observations, taken together with the AC measurements, suggest that the application of DC magnetic fields in this case not only suppresses the quantum tunneling of the magnetization, but also affects the magnetic responses of **1**. However, due to the possible complications of the system involving magnetic anisotropy of Dy(III) ions, the superexchange interactions between the Dy(III) ions through both the TCNQ radicals and hydroxide groups, as well as the inter-chain interactions through the  $\pi$ - $\pi$  stacking interactions between the TCNQ species, it is highly challenging to fully decipher the magnetic properties.

In order to simplify the system by eliminating some of the factors mentioned above, the isostructural compound **2** was synthesized (Figure S2). Given that Gd(III) ions are isotropic ( $S = 7/2$ ,  $g = 2.0$ ), the compound will serve as an excellent model for evaluating the magnetic interactions between the lanthanide ions through TCNQ radicals and hydroxide bridges without the complication of magnetic anisotropy. Under a DC magnetic field of 1000 Oe, the  $\chi T$  values remain nearly constant from  $T = 300$  to 100 K, below which temperatures they decrease gradually down to 1.8 K (Figure S10). The  $\chi T$  value of  $8.3 \text{ emu}\cdot\text{K}\cdot\text{mol}^{-1}$  at 300 K matches well with the presence of one Gd(III) ion and one TCNQ radical (Gd(III),  $S = 7/2$ ,  $g = 2.0$ ; TCNQ radical,  $S = 1/2$ ,  $g = 2.0$ ). Fitting of the experimental data to a Curie-Weiss law  $\chi = C/(T-\theta)$  results in a Curie constant  $C$  of  $8.3 \text{ emu}\cdot\text{K}\cdot\text{mol}^{-1}$  and a Weiss constant  $\theta$  of  $-2.5$  K. The negative Weiss constant is indicative of antiferromagnetic interactions. The temperature dependence of the magnetic susceptibilities was also measured at different DC fields which revealed no variance (Figure S10, inset), indicating that the magnitude of the DC field has no effect on the magnetic interactions.

Two facts have been established by a comparison of the isostructural Gd(III) and Dy(III) compounds, namely that the magnetic field dependent response of the magnetic interactions

in the Dy(III) compound are due to the contribution of magnetic anisotropy, and that antiferromagnetic interactions are dominant in the system. Given these results, the increase of  $\chi T$  at low temperatures and low magnetic fields is likely due to spin-canting of the anisotropic Dy(III) centers, which are coupled by antiferromagnetic interactions through the spin-paired TCNQ radicals and hydroxide bridges.<sup>[13]</sup>

Other possible contributors to the magnetic interactions of the spin centers are inter-chain interactions through the  $\pi$ - $\pi$  stacking of the TCNQ species, in addition to the intra-chain superexchange interactions through the TCNQ radicals and hydroxide bridges. In order to probe the interactions of the stacked TCNQ species, a second isostructural Y(III) compound, **3**, was prepared. As Y(III) is diamagnetic, the only contribution to the magnetic moment is from the paramagnetic TCNQ radicals. Fitting of the experimental data in the high temperature region of  $T = 20\text{-}300$  K to a Curie-Weiss law with a correction for temperature-independent paramagnetism (TIP),  $\chi = C/(T-\theta) + \text{TIP}$ , results in a Curie constant  $C = 0.33 \text{ emu}\cdot\text{K}\cdot\text{mol}^{-1}$ , a Weiss constant  $\theta = -42$  K and a TIP of  $6.8 \times 10^{-4} \text{ emu}\cdot\text{mol}^{-1}$  (Figure S11). The large negative Weiss constant indicates strong antiferromagnetic interactions between the stacked TCNQ radicals, which is likely due to Peierls distortion that results in the spin on TCNQ radicals to pair up at low temperatures.<sup>[14]</sup> The presence of a significant TIP is attributed to Pauli paramagnetism due to the delocalized electrons in the stacked TCNQ species.<sup>[15]</sup> Therefore, we posit that the antiferromagnetic inter-chain interactions through the stacked TCNQ species contribute to the spin-canted states of **1**, in which the Dy(III) ions are coupled antiferromagnetically within the chain. Due to the fact that the magnetic moment contributions from the TCNQ radicals are very small as compared to those of Dy(III), however, the anisotropy of Dy(III) plays a more important role in the magnetic behavior of **1**.



**Figure 5.** Temperature dependence of the electrical conductivity of **1**. Inset: Arrhenius plot of electrical conductivity. The red symbols are experimental data and the black line is the linear fit with an activation energy of 0.43 eV.



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Conductivity measurements were conducted on pressed pellets of polycrystalline samples of **1** by the four-contact (parallel) method. The compound behaves as a semiconductor over the temperature range of  $T = 180\text{--}350\text{ K}$  with a small activation energy of  $0.43\text{ eV}$  (Figure 5). Given that the two stacked bridging TCNQ radicals are in the spin-paired state, it exhibits a good conductivity at room temperature of  $5.0 \times 10^{-4}\text{ S}\cdot\text{cm}^{-1}$  for pressed pellets for which grain boundary effects lead to conductivity that is at least an order or two lower in magnitude than data obtained from single crystal methods.<sup>[9a, 16]</sup>

In summary, a series of isostructural chain compounds,  $\{[\text{Ln}(\text{TPMA})(\mu\text{-TCNQ})(\mu\text{-OH})](\text{TCNQ})_2\cdot\text{CH}_3\text{CN}\}_\infty$  ( $\text{Ln} = \text{Dy}, \text{Gd}, \text{Y}$ ), were synthesized by a facile approach that successfully co-assembles lanthanide ions and partially charged TCNQ radicals. Magnetic and electrical conductivity studies reveal that the Dy(III) compound exhibits slow relaxation of the magnetization at between  $T = 5.0\text{--}8.0\text{ K}$  and semiconducting behavior between over the temperature range of  $180\text{--}350\text{ K}$ . DC magnetic fields suppress the quantum tunneling of the magnetization and also affect the spin-canted antiferromagnetic interactions in **1**. Of course, challenges remain in the realization of improved synergistic interactions between the SMM behavior and the electrical properties which are being tackled in work that is underway.

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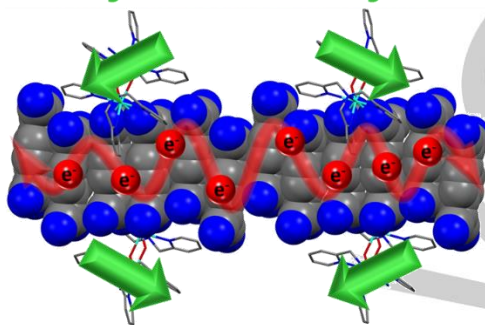
## COMMUNICATION

## Entry for the Table of Contents

Layout 1:

## COMMUNICATION

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**Single-Molecule Magnet****Electrical Conductivity**

Xuan Zhang, Haomiao Xie, Maria Ballesteros-Rivas, Toby J. Woods, and Kim R. Dunbar\*

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Hybrid Dy(III) conducting molecular nanomagnet with partially charged TCNQ radicals