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40. See (*19*) for discussion of the uplift pattern expected from events on a southwest-dipping Sargent-Berrocal system.
41. I thank D. Orange, G. Griggs, G. Valensise, and G. King for discussions. I appreciate critiques of an earlier version of this manuscript by G. Valensise, M. Ellis, and D. Orange, and of later versions by D. Merritts and three anonymous reviewers. Terrace elevations were compiled by J. Buckthal. I thank the donors of the Petroleum Research Fund for partial support.

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A Molecular Ferromagnet with a Curie Temperature of 6.2 Kelvin: $[\mathrm{Mn(C_5(CH_3)_5)_2}]^+[\mathrm{TCNQ}]^-$

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The study of magnetic phase transitions in insulating molecular solids provides new insights into mechanisms of magnetic coupling in the solid state and into critical phenomena associated with these transitions. Only a few such materials are known to display cooperative magnetic properties. The use of high-spin molecular components would enhance intermolecular spin-spin interactions and thus a series of charge-transfer (CT) salts have been synthesized that utilize the spin $S = 1$ molecular cation, $[\mathrm{Mn(C_5(CH_3)_5)_2}]^+$ (decamethylmanganocenium). The structure and cooperative magnetic behavior of $[\mathrm{Mn(C_5(CH_3)_5)_2}]^+[\mathrm{TCNQ}]^-$ (decamethylmanganocenium 7,7,8,8-tetracyano-*p*-quinodimethanide) are reported. This salt is a bulk molecular ferromagnet with the highest critical (Curie) temperature ($T_c = 6.2$ K) and coercive field (3.6×10^3 gauss), yet reported for such a material.

THE SEARCH FOR MAGNETS COMPRISED of discrete molecular components (*1, 2*) represents a new phase in an enterprise that began with the invention of the compass (*3*). However, to date only a small class of ferrimagnets (*4*) and a few well characterized bulk molecular ferromagnets are known: the halo-bis(dialkyl-dichalcogenocarbamato)iron(III) magnets (*5*) and $[\mathrm{Fe(C_5(CH_3)_5)_2}]^+[\mathrm{TCNE}]^-$ (decamethylferrocenium tetracyanoethylenide) (*6*). Furthermore, these latter compounds contain iron, which is ferromagnetic in its elemental form and is the key component of lodestone (magnetite). We report that the donor-acceptor ($[\mathrm{D}]^+[\mathrm{A}]^-$) charge-transfer (CT) salt $[\mathrm{Mn(C_5(CH_3)_5)_2}]^+[\mathrm{TCNQ}]^-$ (I) (decamethylmanganocenium 7,7,8,8-tetracyano-*p*-quinodimethanide) is a true molecular ferromagnet and further propose that molecular ions with $S \geq 1$, such as $[\mathrm{Mn(C_5(CH_3)_5)_2}]^+$, are particularly favorable for the preparation of magnetic molecular materials.

Directed synthesis of insulating ferromagnetic solids from molecular ions requires an understanding of the mechanisms that stabilize the parallel alignment of the component spins. The energy of the ferromagnetic state can be written in the simplest case as a sum of the contributions from pairwise interactions between adjacent spins, $E(F) = -\Sigma J_{ij} \cdot S_i S_j$, where S_i and S_j are the spins of the adjacent interacting sites and J_{ij} is a coupling parameter for the interaction between them (*1, 3, 7–10*). The ground state is ferromagnetic if interactions that cause the spins of individual pairs to align parallel, corresponding to $J_{ij} > 0$, are dominant in three dimensions. Although such interactions include Heisenberg exchange for electrons in orthogonal orbitals (*11*), the most widely disseminated "prescription" (*7*) for generating pairwise ferromagnetic coupling in CT salts is based on a recognition by McConnell (*1*) that we may generalize as follows. Perturbative mixing into the ground state of an interacting $[\mathrm{D}]^+[\mathrm{A}]^-$ pair by a virtual CT excited state of the pair will selectively stabilize a ground state of the same spin multiplicity as the excited state. Thus ferromagnetism is favored by constituents for which the total spin angular momentum (S_T) of the lowest-energy virtual CT state of an interacting $\mathrm{D^+\ A^-}$ pair corresponds to the sum of the spins of $\mathrm{D^+}$ and $\mathrm{A^-}$, $S_T = S(\mathrm{D^+}) + S(\mathrm{A^-})$; antiferromagnetism (or ferrimagne-

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tism) is favored if $S_T = |S(D^+) - S(A^-)|$. Miller and Epstein proposed that the three-dimensional (3-D) ferromagnetic ground state of the mixed-stack linear chain CT salt they prepared, $[Fe(C_5(CH_3)_5)_2]^+[TCNE]^-$ (*6*), is stabilized by this mechanism (*7, 8*). Further attempts to achieve bulk ferromagnetism by preparing [metallocenium]$^+$[A$^-$] CT salts where the anions as well as the cations have $S = 1/2$ have been less successful: several salts displaying ferromagnetic coupling have been prepared but no other 3-D ferromagnet has been reported (*8, 12–14*).

We reasoned that the use of cation-anion components with spin $S \geq 1$ would enhance bulk magnetism in a CT salt. According to the equation for $E(F)$, taking either $S(D^+)$ or $S(A^-)$ or both ≥ 1, rather than equal to ½, would lower the energy of the ferromagnetic ground state for a given J_{ij} when ferromagnetic interactions dominate. In addition, components with $S > 1/2$ will increase the saturation moment, M_s, for the state with all spins ferromagnetically aligned: $M_s = N\beta[g_D S(D^+) + g_A S(A^-)]$, where N is Avogadro's number, β is the electronic Bohr magneton, and $g_{D/A}$ is the appropriate Lande factor. Within the McConnell mechanism, ferromagnetism requires that the key CT state or states have the correct spin to give $J_{ij} > 0$. However, even if the D^+ and A^- spins align antiferromagnetically, a net moment still would remain when $S(D^+) \neq S(A^-)$, and the resultant material could be a bulk magnet, namely a ferrimagnet. In this case, the saturation magnetization would be $M_s = N\beta|g_D S(D^+) - g_A S(A^-)|$. Finally, the possible interplay between the intermolecular coupling among the spins and the intramolecular zero-field splittings for a component with $S > 1/2$ can lead to unusual magnetic characteristics (*5*).

Within this framework, $[D]^+ = [Mn(C_5(CH_3)_5)_2]^+$ (decamethylmanganocenium) is an excellent choice as the cation for several reasons. (i) The parent donor, $[Mn(C_5(CH_3)_5)_2]$, is easily oxidized to $[Mn(C_5(CH_3)_5)_2]^+$ ($E_{+/0} = -0.56$ V versus the standard calomel electrode) (*15*) and thus will form CT salts with a wide variety of acceptors. (ii) $[Mn(C_5(CH_3)_5)_2]^+$ has spin $S = 1$ (*15*). (iii) In a salt with this cation and a paramagnetic anion (A−; $S = 1/2$) derived from a strong acceptor, the lowest energy virtual CT state would be $[Mn(C_5(CH_3)_5)_2]^{2+}[A]^{2-}$. This compound we expected to have $S_T = S(D^{2+}) = 3/2$, which would lead to a pairwise ferromagnetic interaction ($J_{ij} > 0$) for an A^- that has $S = 1/2$. The expectation is based on the observation that D^{2+} formally contains Mn^{4+} (d^3), which would have $S = 3/2$, whereas A^{2-} would be diamagnetic if A^- is orbitally nondegenerate. (iv) Finally, the absence of the element Fe removes the possibility that decomposition could lead to contamination by ferromagnetic metallic Fe because metallic Mn is antiferromagnetic.

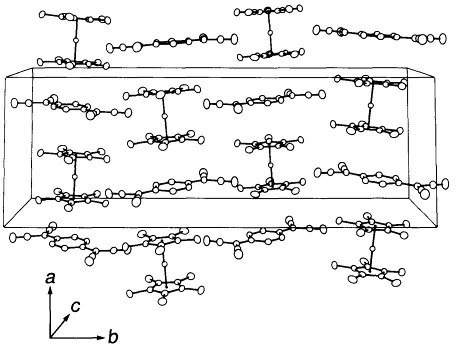

Fig. 1. Packing diagram of **I** showing a perspective view of the . . .$D^+A^-D^+A^-$. . . stacking arrangement in the *bc* plane.

We have synthesized a series of molecular CT salts with the $S = 1$ cation $[Mn(C_5(CH_3)_5)_2]^+$ and have found that it does indeed support ferromagnetic interactions in a broad class of compounds (*16*). In particular, **I** undergoes a magnetic phase transition to a 3-D ordered ferromagnetic state at 6.2 K, the highest temperature yet reported for a molecular ferromagnet. Compound **I** crystallizes as lustrous, dark, parallelpipeds in the monoclinic space group $P2_1/n$, with four formula units per unit cell. The structure of **I**, as solved by single-crystal x-ray diffraction techniques, consists of . . .$D^+A^-D^+A^-$. . . stacks in which $Mn(C_5(CH_3)_5)_2^+$ (D^+) and TCNQ$^-$ (A^-) ions alternate along *a* (Fig. 1). In addition, the closest interstack contacts also are between D^+ and A^- (*17, 18*). A similar stack structure has been observed in other decamethylmetallocenium CT salts (*8, 13*). The pentamethylcyclopentadienyl rings of the cation in **I** adopt an eclipsed conformation ($\sim D_{5h}$ local symmetry), not the staggered conformation previously observed in the structure of the decamethylmanganocene (*19*). Compound **I** is isomorphous with the structure first reported (*20*) for the Fe analog, $[Fe(C_5(CH_3)_5)_2]^+[TCNQ]^-$. However, detailed comparisons are not possible. The structure reported for the Fe compound was that of a crystal in which extended exposure to the atmosphere had caused the TCNQ anions to undergo a solid-state reaction with O_2 (or H_2O) to produce the α,α-dicyano-*p*-toluoylcyanide anion. X-ray powder diffraction data indicated that the freshly prepared Fe compound is isomorphous to the exposed material (*21*).

The magnetic susceptibility, χ_m, of polycrystalline **I** was measured with a superconducting quantum interference device (SQUID) magnetometer. The temperature dependencies of $\chi_m T$ and χ_m^{-1} recorded at 500 G are shown in Fig. 2. The room temperature value, $\chi_m T = 2.44 \pm 0.01$ cm^3 K mol^{-1}, is greater than the spin-only value (1.38 cm^3 K mol^{-1}) calculated for an uncorrelated two-spin system with $S_D = 1$ and $S_A = 1/2$ when a spin-only value of $g = 2$ is used for both spins. Because organic radicals such as TCNQ$^{\cdot-}$ display spin-only moments, the high value of $\chi_m T$ probably is due to an orbital contribution to the *g*-factor of $[Mn(C_5(CH_3)_5)_2]^+$. The value of $\chi_m T$ for **I** is constant from ambient to ~50 K, and a plot of χ_M^{-1} versus T is linear (Fig. 2). Together these results show that the susceptibility of **I** follows the Curie-Weiss law, $\chi_m = C/(T - \Theta)$ for $T \gtrsim 50$ K. The Weiss parameter, which corresponds to the T-intercept of χ_M^{-1}, is $\Theta = +10.5 \pm 0.5$ K. Its positive sign indicates that ferromagnetic interactions ($J_{ij} > 0$) dominate. If it is assumed that the largest exchange parameter is associated with near neighbors along a chain, then it can be shown that $J_{D^+A^-}/k = (11/8)\Theta = 14$ K (*10*). As the sample is cooled below 20 K, an abrupt increase in $\chi_m T$ signals the approach of a ferromagnetic phase transition. Below the transition region, $T \lesssim 5$ K, the magnetization of **I** becomes saturated: χ_m is roughly constant, and $\chi_m T$ decreases linearly with T.

The occurrence of a ferromagnetic phase

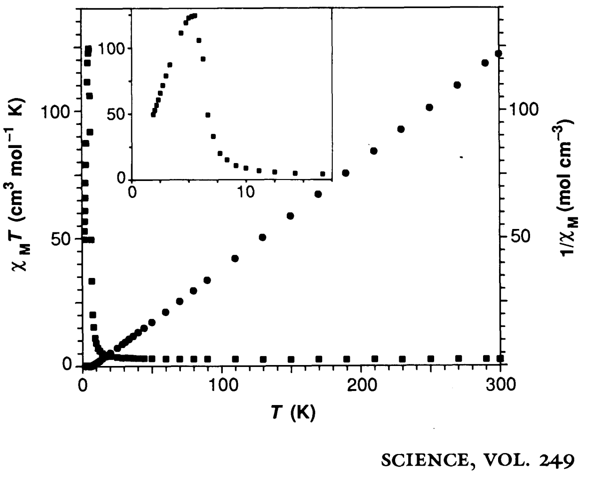

Fig. 2. Temperature dependence of $\chi_m T$ (squares) and χ_m^{-1} (circles) for **I** recorded at 500 ± 10 G. Inset: expansion of the 1.9 to 25 K region of the $\chi_m T$ plot. The susceptibility plotted here has been corrected for core diamagnetism as calculated from Pascal's constants.

transition was confirmed by measuring the magnetization M of **I** as a function of temperature and field in the ranges of 1.9 to 10 K and 0 to 50,000 G. As **I** is cooled in a low field (50 G), its magnetization (Fig. 3) shows a sharp transition, increasing abruptly between 6.5 and 6 K. The critical temperature T_c is taken at the maximum of the slope dM/dT: $T_c = 6.2 \pm 0.1$ K. At temperatures below T_c ($T <$ 4 to 5 K), the magnetization saturates to a constant value (Fig. 3), as expected for a bulk ferromagnet. The field dependence of the magnetization at 3 K (Fig. 4) is characteristic of a polycrystalline 3-D ferromagnet. The magnetization climbs steeply to a value of $\sim 1.35 \times 10^4$ cm^3 G mol^{-1} by 1000 G, and then gradually increases to a maximum of $1.67 \pm 0.01 \times 10^4$ cm^3 G mol^{-1} at the highest field attainable, 50,000 G. This result shows that **I** is indeed a ferromagnet, not a ferrimagnet, which would display a much smaller saturation magnetization. The expression for M_s of a ferrimagnet (see above) gives $M_s = 5.58 \times 10^3$ cm^3 G mol^{-1} when $S(D^+) = 1$, $S(A^-) = 1/2$, and $g = 2$ for both spins. Even a significant orbital contribution to the g factor of $[Mn(C_5(CH_3)_5)_2]^+$ could not increase M_s to give agreement with the value observed for polycrystalline **I**.

The zero-field susceptibility $(dM/dH)_{H=0}$ is extremely large but not infinite, and M is not a constant for fields above ~1000 G (Fig. 4), in contrast to what would be observed in the limit of $T << T_c$ for a perfect ferromagnet aligned along the easy axis. These features of Fig. 4 arise because of demagnetization effects and because the susceptibility is averaged over all orientations of the polycrystalline material, including both easy and hard magnetization axes. Indeed, preliminary single-crystal studies indicate that the magnetization of **I** is exceptionally anisotropic.

Definitive verification that **I** is a bulk ferromagnet is given by the observation that at temperatures below T_c its magnetization exhibits hysteretic behavior upon cycling the applied field; the major hysteresis loop at 2 K is shown in the inset to Fig. 4. The remnant magnetization, the value at zero applied field, is large, $1.37 \pm 0.01 \times 10^4$ cm^3 G mol^{-1}, and is equal to that expected from an extrapolation to zero field of the high-field portion of the saturation magnetization curve (Fig. 4). The coercive field, defined (*10*) as that where $M = 0$ (Fig. 4, inset), is $3.6 \pm 0.1 \times 10^3$ G, the largest yet reported for a molecular ferromagnet or ferrimagnet.

These measurements demonstrate that **I** is a molecular ferromagnet with excellent crystallographic characteristics that make it well suited for further detailed examination. In addition, these results support the use of high-spin ($S \geq 1$) components in CT salts to broaden the class of magnetically ordered molecular solids. Thus of the two isomorphous compounds, $[M(C_5(CH_3)_5)_2]^+[TCNQ]^-$ with M either Fe or Mn, the salt previously prepared with the $S(D^+) = 1/2$ $[Fe(C_5(CH_3)_5)_2]^+$ ion (*20*) has an antiferromagnetic ground state and displays metamagnetic behavior, with a critical field of 1.6 kG and a Neel temperature of 2.55 K. In contrast, the new salt prepared with $S(D^+) = 1$ $[Mn(C_5(CH_3)_5)_2]^+$ has a 3-D ferromagnetically ordered ground state and both a high critical temperature (T_c = 6.2 K) and coercive field (3.6×10^3 G) for a molecular ferromagnet.

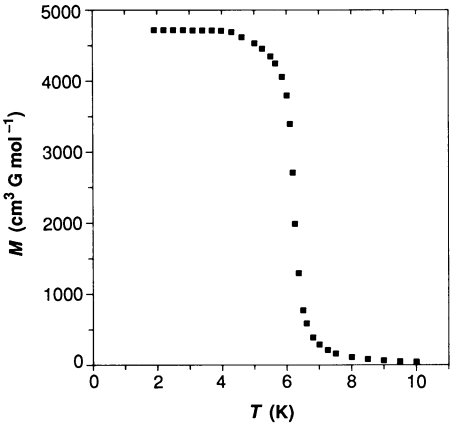

Fig. 3. Field-cooled magnetization M of **I** in an external field of 50 ± 1 G.

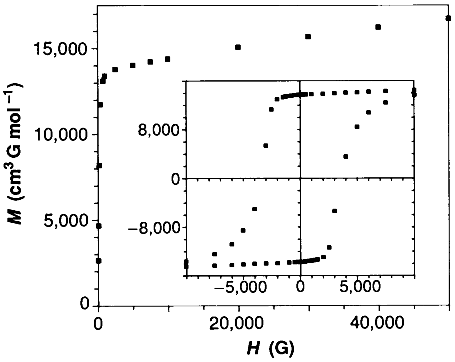

Fig. 4. Field (H) dependence of the magnetization for polycrystalline **I** recorded at 3 K. The inset shows a hysteresis loop, $M = f(H)$ for **I** recorded at 2 K.

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17. We prepared **I** by combining solutions of $[Mn(C_5(CH_3)_5)_2]^+[PF_6]^-$ (*15*) and NH_4TCNQ (*18*). The composition, as well as the absence of Fe impurities, was verified by elemental analysis. Crystallographic data for **I**: monoclinic, space group $P2_1/n$ (C_{2h}, no. 14); $a = 10.829 \pm 0.002$ Å, $b = 31.014 \pm 0.004$ Å, and $c = 8.544 \pm 0.001$ Å, $\beta = 103.39 \pm 0.02°$, $V = 2792 \pm 1$ Å^3, $Z = 4$. The structure was solved by a combination of direct methods and Fourier syntheses in TEXSAN 4.0. Full matrix least-squares refinement gave the final agreement factors, $R(F) = 0.034$, and $R_w(F) = 0.043$ for 3483 reflections [with $F^2 \geq 3\sigma(F^2)$] measured between $4 \leq 2\theta \leq 50$ at −120°C (Mo Kα radiation, $\lambda = 0.71069$). Full interstack interactions and other structural details of **I** will be discussed later [W. E. Broderick, J. A. Thompson, M. Sabat, E. P. Day, B. M. Hoffman, in preparation]. The atomic coordinates for this work are available on request from the Director of the Cambridge Crystallographic Data Centre, University Chemical Laboratory, Lensfield Road, Cambridge, CB2 1EW, England. Requests should be accompanied by the full literature citation for this communication.
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21. A subsequent single-crystal structural determination of $[Fe(C_5(CH_3)_5)_2]^+[TCNQ]^-$ found that the salt crystallized in a different space group, triclinic $P\bar{1}$; a $\ldots D^+A^-D^+A^- \ldots$ stack arrangement was also observed in this second structure [J. S. Miller *et al.*, *J. Phys. Chem.* **91**, 4344 (1987)].
22. Supported by the Solid State Chemistry Program of the National Science Foundation (grant DMR-8818599) and by the Northwestern University Materials Research Center under the NSF-MRL program (grant DMR-8821571). We thank the research group of L. Que, Jr., for technical assistance.

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