Electronic Supporting Information

Metamagnetism in a π-stacked *bis*-dithiazolyl radical

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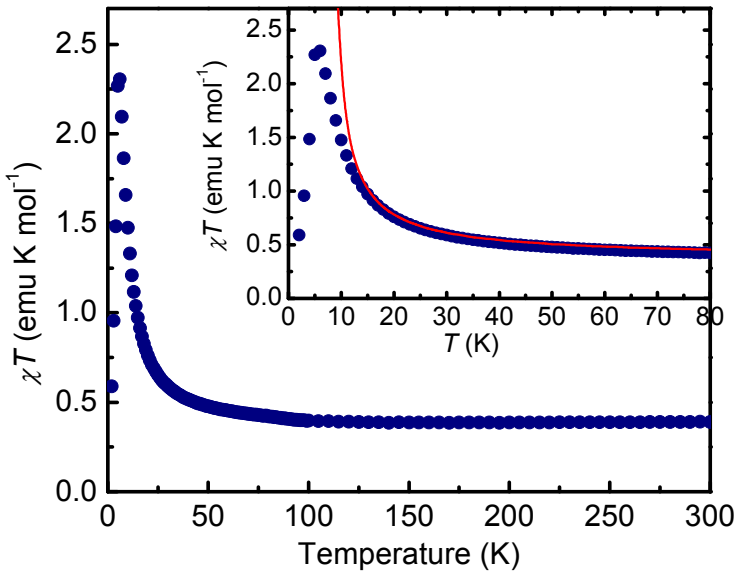
Figure S1. Fitting of the Magnetic Data for **1a**.

The low temperature (T = 10-80 K) magnetic susceptibility of **1a** was fitted (Equation 1) to the Baker 1D S = ½ FM chain model[1] modified to include a molecular field parameter (zJ') to account for interchain interactions (Equation 2). Contributions from Temperature Independent Paramagnetism (TIP) and Curie-Weiss impurities (α) were also included (Equation 3).

$$\chi_{1D} = \frac{Ng^2\beta^2}{4k(T-\theta)}\left[\frac{1.0 + 5.7979916X + 16.902653X^2 + 29.376885X^3 + 29.832959X^4 + 14.036918X^5}{1.0 + 2.7979916X + 7.0086780X^2 + 8.653644X^3 + 4.5743114X^4}\right]^{2/3} \quad \text{(Eqn 1)}$$

$$where\ X = \frac{|J|}{2kT}$$

$$\chi_{2D} = \frac{\chi_{1D}}{1 - \frac{2(zJ')\chi_{1D}}{Ng^2\beta^2}} \quad \text{(Eqn 2)} \qquad \chi_{calc} = \left\{\frac{\alpha\left[Ng^2\beta^2 S(S+1)\right]}{3kT} + TIP + \left[1-\alpha\right]\chi_{2D}\right\} \quad \text{(Eqn 3)}$$



Baker Fit to data from 10 - 80 K:

J = +6.17 cm^{-1}, $z\boldsymbol{J}'$ = +4.90 cm^{-1}, g = 2.008, θ = 0.00 K, α = 0.000, TIP = 0.0001 emu mol^{-1}, $R(\chi)$ = 0.0279.*

*$R(\chi) = [\Sigma(\chi_{obs} - \chi_{calc})^2/\Sigma(\chi_{obs})^2]^{½}$

[1]G. A. Baker, G. S. Rushbrooke and H. E. Gilbert, *Phys. Rev.*, 1964, **135**, A1272.

Table S1. Calculated Exchange Energies for **1a**.

Triplet and broken symmetry singlet (guess=mix) energies were derived from single point calculations, performed using Gaussian 03W[2] at the UB3LYP/6-311G(d,p) level, with SCF = tight. Atomic coordinates were taken from crystallographic data at T = 35 K, 100 K and 293 K.

$$J = \frac{-(E_{TS} - E_{BSS})}{< S^2 >_{TS} - < S^2 >_{BSS}}$$

35K Data	Distance	Triplet (H)	<S2>TS	BS Singlet (H)	<S2>BSS	J (cm-1)
J-pi	*a*	-3976.23200484	2.0658	-3976.23197128	1.0648	7.36
J1	d1	-3976.23434226	2.0658	-3976.23431841	1.0656	5.23
J2	d2	-3976.23460916	2.0651	-3976.23461072	1.0634	-0.34
J2'	d2'	-3976.23535937	2.066	-3976.23536298	1.066	-0.79
J3	d3	-3976.23493400	2.0655	-3976.23492227	1.0654	2.57
J3'	d3'	-3976.23489758	2.0658	-3976.23489514	1.0655	0.54

100K Data	Distance	Triplet (H)	<S2>TS	BS Singlet (H)	<S2>BSS	J (cm-1)
J-pi	*a*	-3976.26681602	2.0624	-3976.26680431	1.0601	2.56
J1	d1	-3976.26921639	2.0625	-3976.26919188	1.0624	5.38
J2	d2	-3976.26950498	2.0619	-3976.26950888	1.0602	-0.85
J2'	d2'	-3976.26998528	2.0628	-3976.26999009	1.0627	-1.06
J3	d3	-3976.26955878	2.0623	-3976.26954789	1.0622	2.39
J3'	d3'	-3976.26973302	2.0626	-3976.26973487	1.0621	-0.41

293K Data	Distance	Triplet (H)	<S2>TS	BS Singlet (H)	<S2>BSS	J (cm-1)
J-pi	*a*	-3976.14485469	2.0655	-3976.14482875	1.0637	5.68
J1	d1	-3976.14576298	2.0658	-3976.14574174	1.0656	4.66
J2	d2	-3976.14580607	2.065	-3976.14580435	1.0636	0.38
J2'	d2'	-3976.14619568	2.0658	-3976.14619810	1.0658	-0.53
J3	d3	-3976.14597107	2.0655	-3976.14596017	1.0655	2.39
J3'	d3'	-3976.14598720	2.0658	-3976.14598843	1.0654	-0.27

[2]Gaussian 03, Revision C.02:
M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, J. A. Montgomery, Jr., T. Vreven, K. N. Kudin, J. C. Burant, J. M. Millam, S. S. Iyengar, J. Tomasi, V. Barone, B. Mennucci, M. Cossi, G. Scalmani, N. Rega, G. A. Petersson, H. Nakatsuji, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, M. Klene, X. Li, J. E. Knox, H. P. Hratchian, J. B. Cross, V. Bakken, C. Adamo, J. Jaramillo, R. Gomperts, R. E. Stratmann, O. Yazyev, A. J. Austin, R. Cammi, C. Pomelli, J. W. Ochterski, P. Y. Ayala, K. Morokuma, G. A. Voth, P. Salvador, J. J. Dannenberg, V. G. Zakrzewski, S. Dapprich, A. D. Daniels, M. C. Strain, O. Farkas, D. K. Malick, A. D. Rabuck, K. Raghavachari, J. B. Foresman, J. V. Ortiz, Q. Cui, A. G. Baboul, S. Clifford, J. Cioslowski, B. B. Stefanov, G. Liu, A. Liashenko, P. Piskorz, I. Komaromi, R. L. Martin, D. J. Fox, T. Keith, M. A. Al-Laham, C. Y. Peng, A. Nanayakkara, M. Challacombe, P. M. W. Gill, B. Johnson, W. Chen, M. W. Wong, C. Gonzalez, and J. A. Pople, Gaussian, Inc., Wallingford CT, 2004.