ELECTRONIC SUPPLEMENTARY INFORMATION

for

A co-crystal of 2-(1'-pyrenyl)-4,4,5,5-tetramethyl-4,5-dihydro-1H-imidazole-3-oxide-1-oxyl with octafluoronaphthalene

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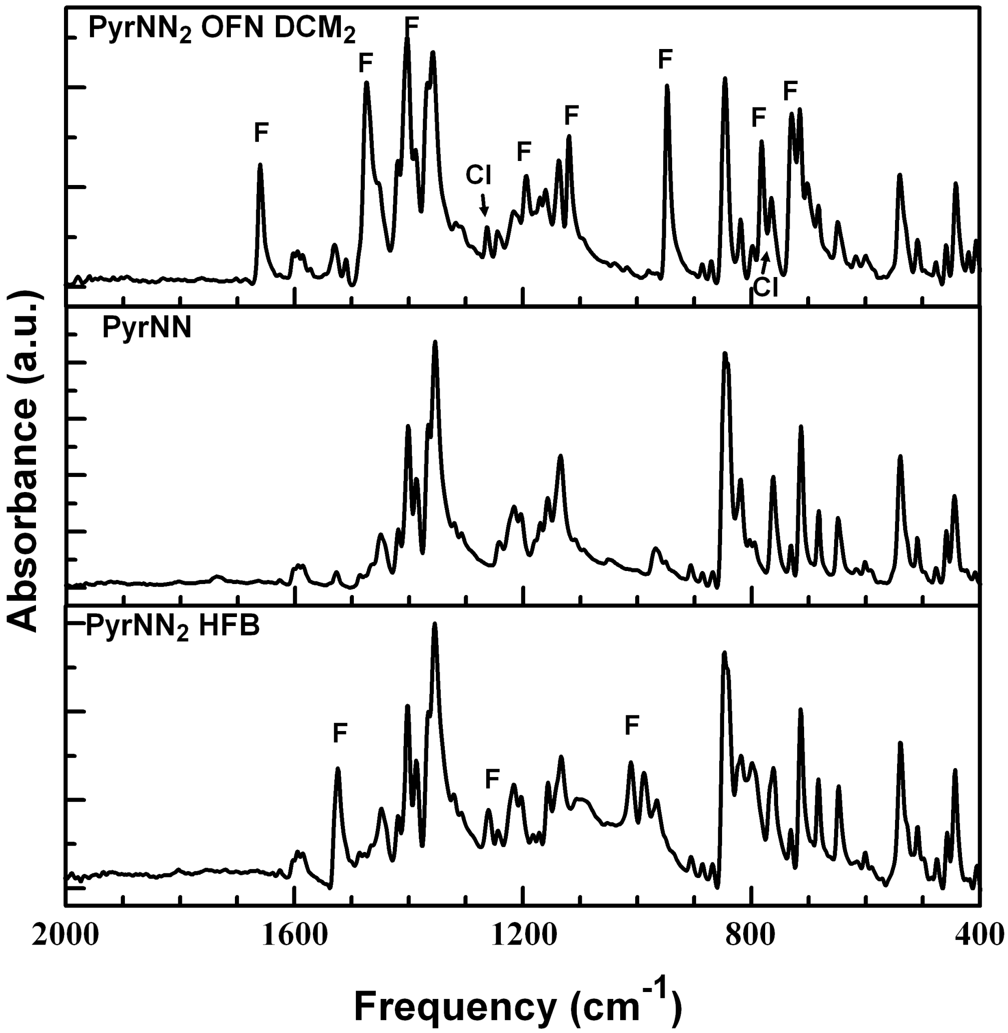
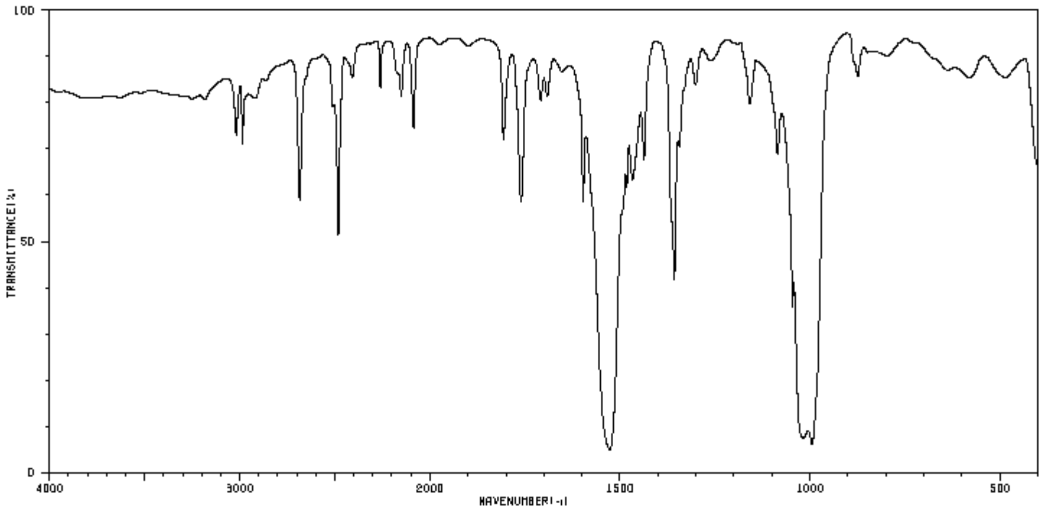
Figure S1. Neat solid ATR-FTIR spectra of $PyrNN_2 \cdot OFN \cdot DCM_2$, PyrNN, $PyrNN_2 \cdot HFB$. (F = fluoroarene component, Cl = DCM = dichloromethane); see also literature spectra for HFB, OFN, DCM on pages S2-S3.



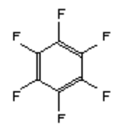
Spectral assignments for peaks above labled F and Cl are based on spectra for OFN. HFB, DCM from the Spectral Database for Organic Compounds, SDBS. a free site organized by National Institute of Advanced Industrial Science and Technology (AIST), Japan. Access made at http://riodb01.ibase.aist.go.jp/sdbs/ (National Institute of Advanced Industrial Science and Technology, 7 Aug 2012).

See spectra from the SDBS site on the following two pages.

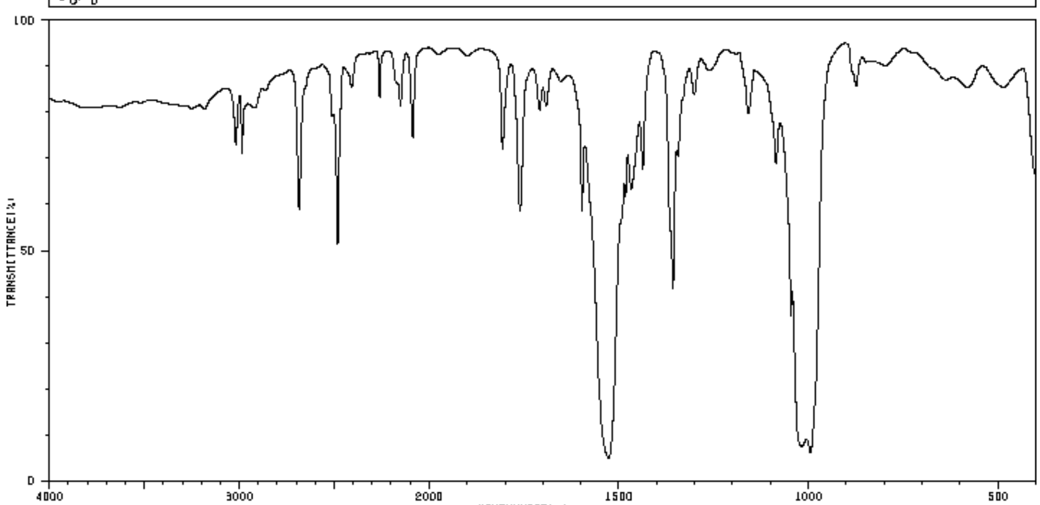
HIT-NO=2973	SCORE= ( )	SDBS-NO=5441	IR-NIDA-09704 : LIQUID FILM
HEXAFLUOROBENZENE			
C_6F_6			



3184	77	2261	79	1692	79	1364	62	1018	7
3018	70	2171	84	1653	84	1358	39	1005	8
2985	86	2150	79	1597	57	1344	68	994	6
2684	57	2086	72	1527	4	1301	81	886	86
2509	77	1806	70	1483	58	1159	77	874	81
2481	50	1760	57	1467	80	1085	65	580	61
2407	81	1709	77	1437	66	1046	36	484	81



HIT-NO=2973	SCORE= ( )	SDBS-NO=5441	IR-NIDA-09704 : LIQUID FILM
HEXAFLUOROBENZENE			
C_6F_6			



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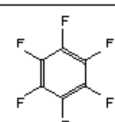
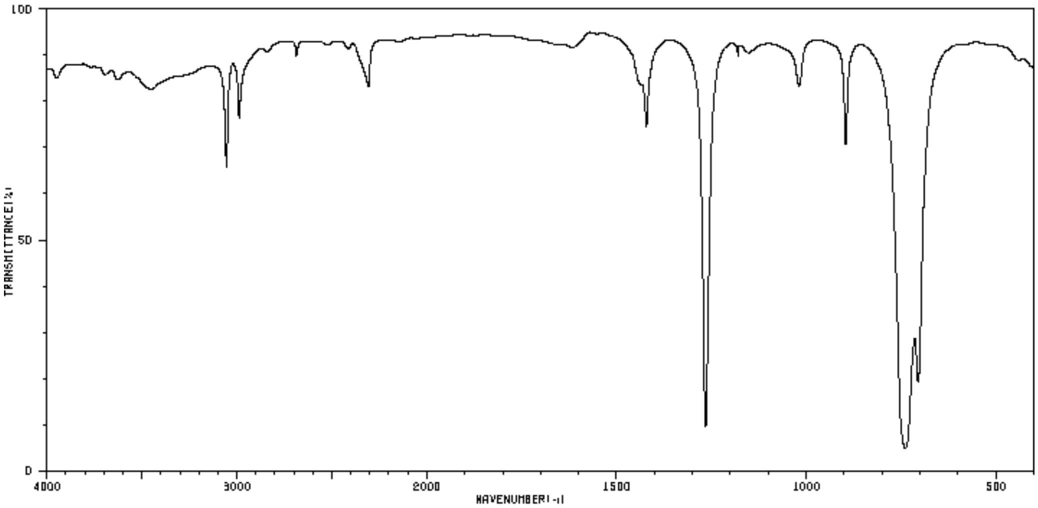
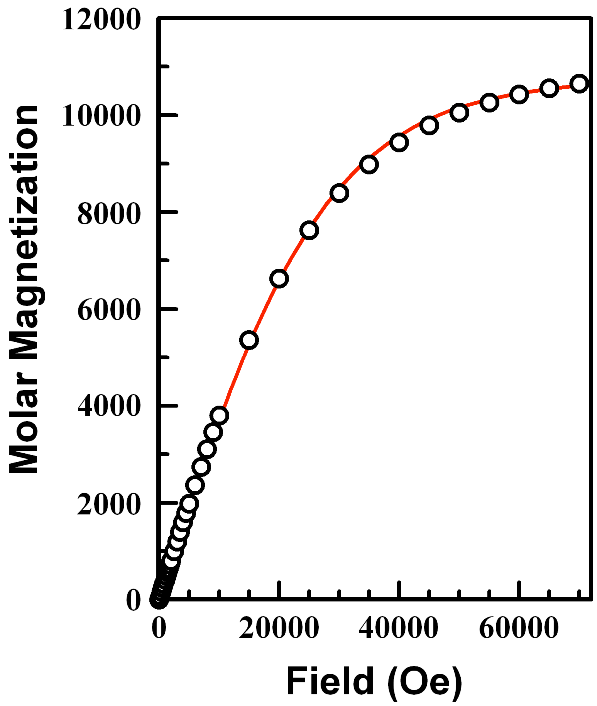


HIT-NO=1231	SCORE= ( )	SDBS-NO=891	IR-NIDA-05334 : LIQUID FILM
DICHLOROMETHANE			
CH_2CL_2			



3949	81	2307	79	896	68
3627	81	1433	81	739	4
3455	79	1422	72	706	18
3446	79	1266	9		
3055	64	1181	86		
2987	74	1028	84		
2687	86	1019	78		

Figure S2. Magnetization versus field data for $PyrNN_2 \cdot OFN \cdot DCM_2$ at 1.8 K, with Brillouin fitting function for S = 1/2.



For the theoretical Brillouin curve of magnetization versus field at 1.8 K (solid red line), the following equation was used

$$M = Ng\mu_B \cdot S \cdot B_S(x)$$
$$B_S(x) = \frac{2S+1}{2S}\coth\left(\frac{2S+1}{2S}x\right) - \frac{1}{2S}\coth\left(\frac{x}{2S}\right)$$
$$x = g\mu_B H / k_B T$$

where S = 1/2 and g = 1.9 were used to fit the observed data. In the equation, M is the molar magnetization, H is external magnetic field, T is temperature, and other terms are constants with the usual meanings

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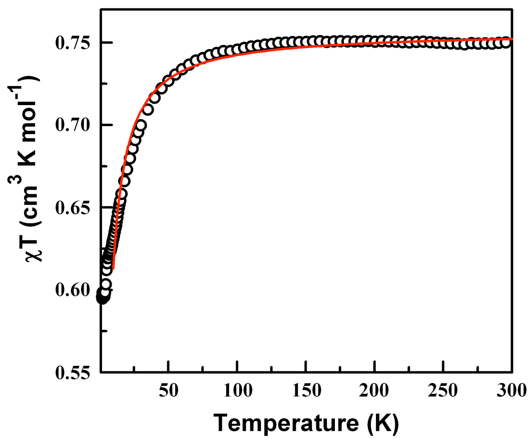
Figure S3. χT vs T data with fitting to 1-D chain model for $PyrNN_2 \cdot OFN \cdot DCM_2$ at 1000 Oe (dc).

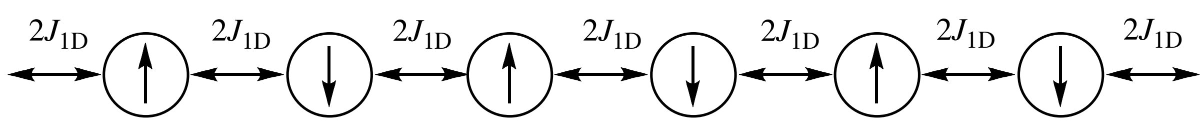


1-D linear Heisenberg chain model for S =1/2 spin units

From Swank, D. D.; Landee, C. P., Willet, R. D. *Phys. Rev. B*, **1979**, *20*, 2154.

$$\chi T = \frac{Ng^2\beta^2}{4k} \cdot \frac{T}{T-\theta} \cdot \left[\frac{1 + A\cdot\left(J/2kT\right) + B\cdot\left(J/2kT\right)^2 + C\cdot\left(J/2kT\right)^3 + D\cdot\left(J/2kT\right)^4 + E\cdot\left(J/2kT\right)^5}{1 + F\cdot\left(J/2kT\right) + G\cdot\left(J/2kT\right)^2 + H\cdot\left(J/2kT\right)^3 + I\cdot\left(J/2kT\right)^4} \right]^{2/3} \qquad H = -2J\sum_{N=1}^{\infty} S_1 \cdot S_{1+N}$$

A = 5.7979916, B = 16.902653, C = 29.376885, D = 29.832959, E = 14.036918
and F = 2.7979916, G = 7.0086780 , H = 8.6538644, I = 4.5743114



This fit was done for data where T > 10 K; the fitted results are J/k = (–)0.96±0.03 K, g = 2.0093±0.002.