

affected by cooling to liquid air temperature. The infra-red spectra in the region 200 to 3000 cm^{-1} have been measured and are referred to in the discussion.

The pressure dependence of the magnetic susceptibility of the *n*-butyl complex has been measured in chloroform solution. The susceptibility of the solvent does not change with pressure, nor does that of a typical 'magnetically normal' complex

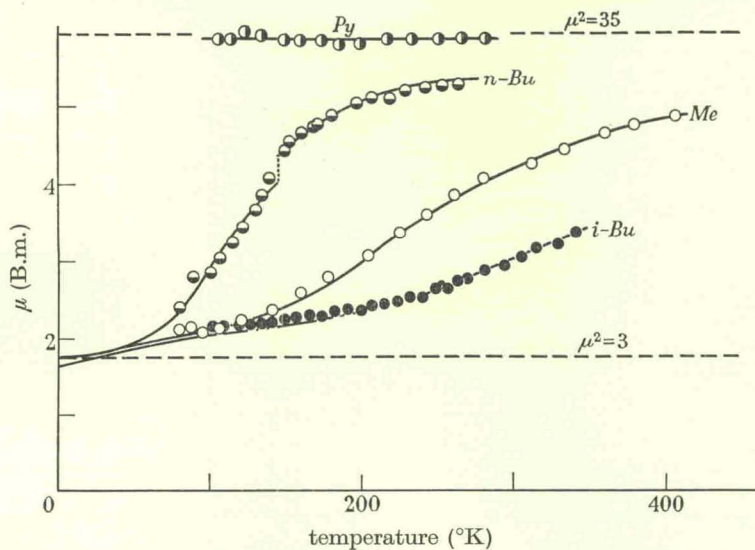


FIGURE 6. Variation of the magnetic moments of $[\text{Fe}(\text{S}_2\text{CNR}_2)_3]$ with temperature. The full curves are calculated from equations (1), as modified by (2) and (4). *Me*:*R* = methyl; *n-Bu*:*R* = *n*-butyl; *i-Bu*:*R* = *i*-butyl; *Py*:*NR*₂ = pyrrolidyl.

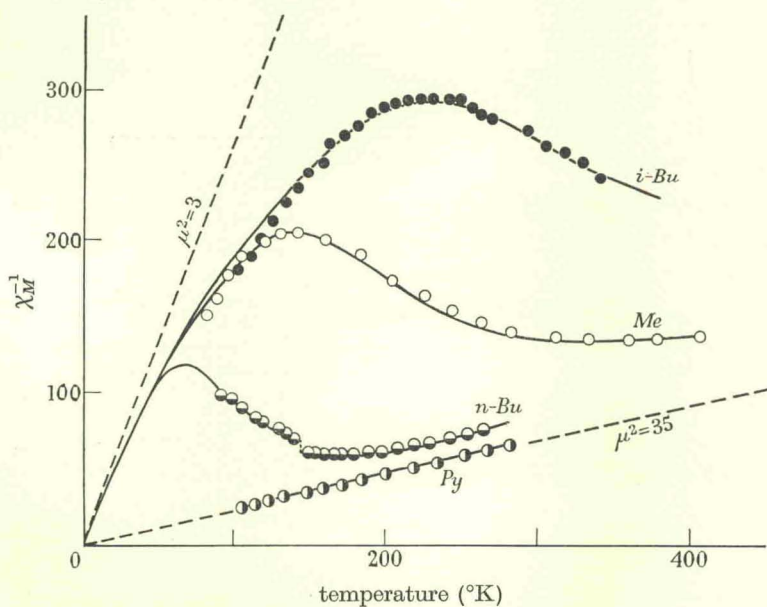


FIGURE 7. Variation of the χ_M^{-1} of $[\text{Fe}(\text{S}_2\text{CNR}_2)_3]$ with temperature. See also legend to figure 6.

(NiCl_2 in water). The susceptibility of the iron (III) dithiocarbamate complex, on the other hand, decreases dramatically with increased pressure (figure 9).

The highly desirable paramagnetic resonance studies have only recently been instituted by Mr H. F. Symmons of the National Standards Laboratory, C.S.I.R.O., Sydney, but meanwhile we note that all the observations so far made are in qualitative accord with those predicted for a d^5 complex in the crossover region.

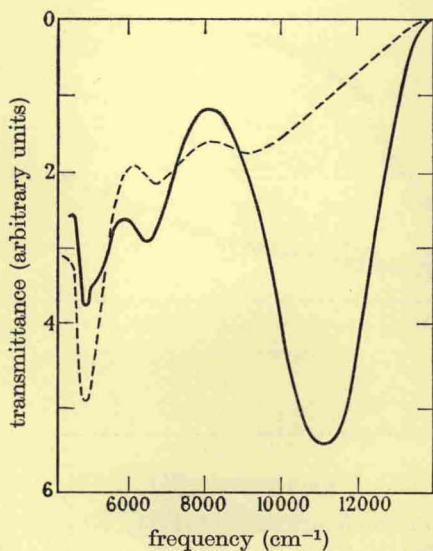


FIGURE 8. Absorption spectra of $[\text{Fe}(\text{S}_2\text{CNMe}_2)_3]$ in KCl (pressed disk); long wavelength region. Full line: 90 °K. Broken line: room temperature.

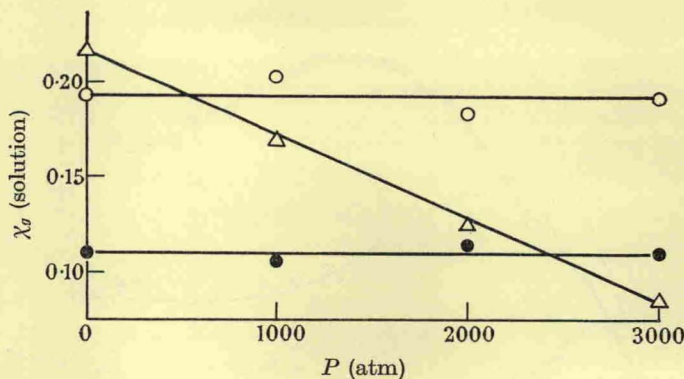


FIGURE 9. Pressure dependence of magnetic susceptibility. ○, NiCl_2 in water ($\chi_{\text{soln}} - 1.20$), ●, CHCl_3 ($\chi_g + 0.60$), Δ , $[\text{Fe}(\text{S}_2\text{CN}n\text{-Bu}_2)_3]$ in CHCl_3 (χ_{soln}).

DISCUSSION

It is clear that the compounds presented here reproduce qualitatively all the requirements which we have formulated for crossover behaviour. Quantitative comparisons between experiment and expectation follow.