low- or high-spin behaviour ($E/\zeta = \pm\infty$) are seen only when $|E|/\zeta < 1$. The resulting curves are not, however, notably different in shape from those obtained in other situations where μ is temperature dependent. In this connexion it is worth remarking that one is likely, in practice, to be able to observe a portion only of the $\mu(T)$ curve, since thermal decomposition and phase changes invariably impose an upper

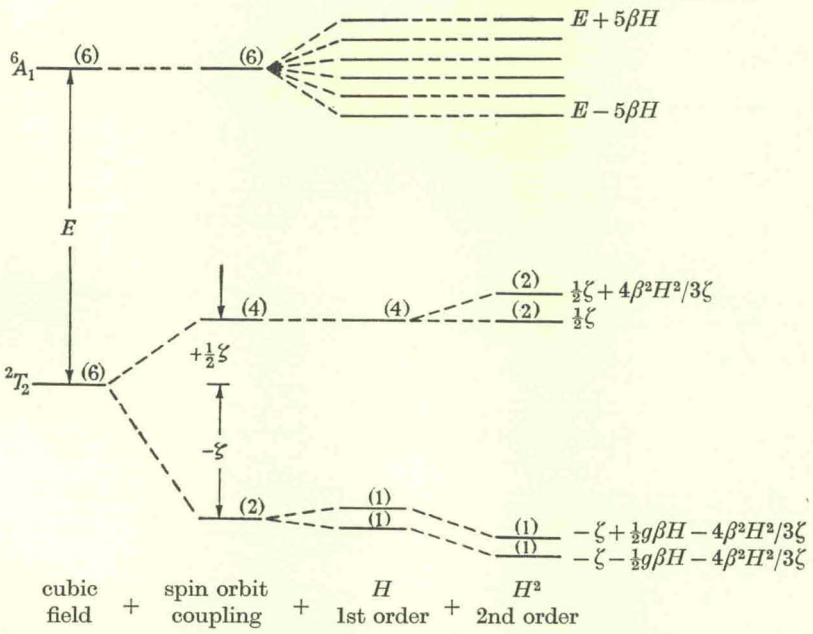

FIGURE 3. Energy levels (not to scale) of configuration d^5 in the crossover region.

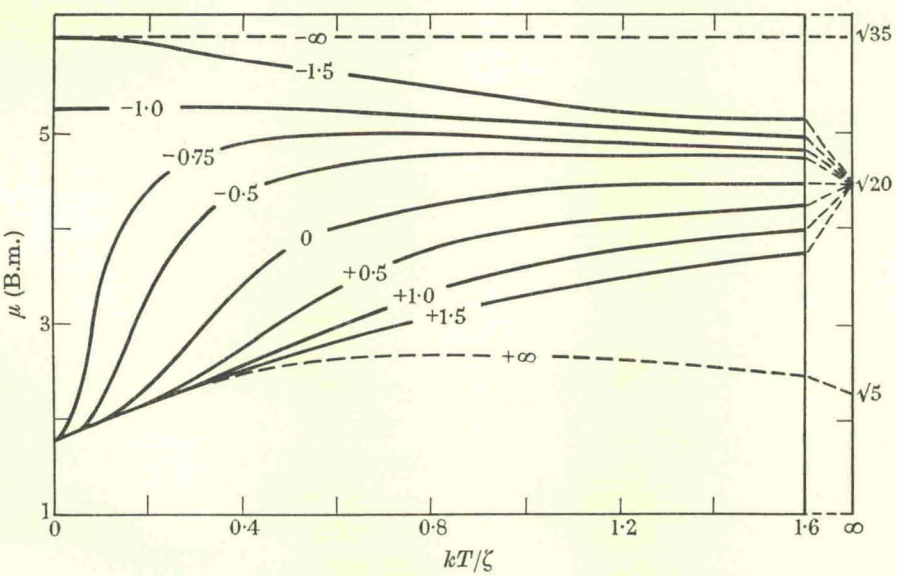

FIGURE 4. Calculated values of the effective magnetic moment μ, assuming $g = 2$, and values of E/ζ as given on the curves.

temperature limit on measurements. Further, which portion of the $\mu(T)$ curve is being seen is also uncertain, since ζ is not known *a priori*; ζ can only be regarded as a parameter whose value probably lies somewhere between 300 and 440 cm^{-1} (see Figgis 1961).

The exceptional nature of the magnetic behaviour is better revealed by the temperature dependence of the reciprocal molar susceptibility, $\chi_M^{-1} = 3kT/N\beta^2\mu^2$. From figure 4 are derived the full-line curves of χ_M^{-1} in figure 5. As Guha (1963) has pointed out, under certain conditions a $\mu(T)$ dependence of the kind shown in

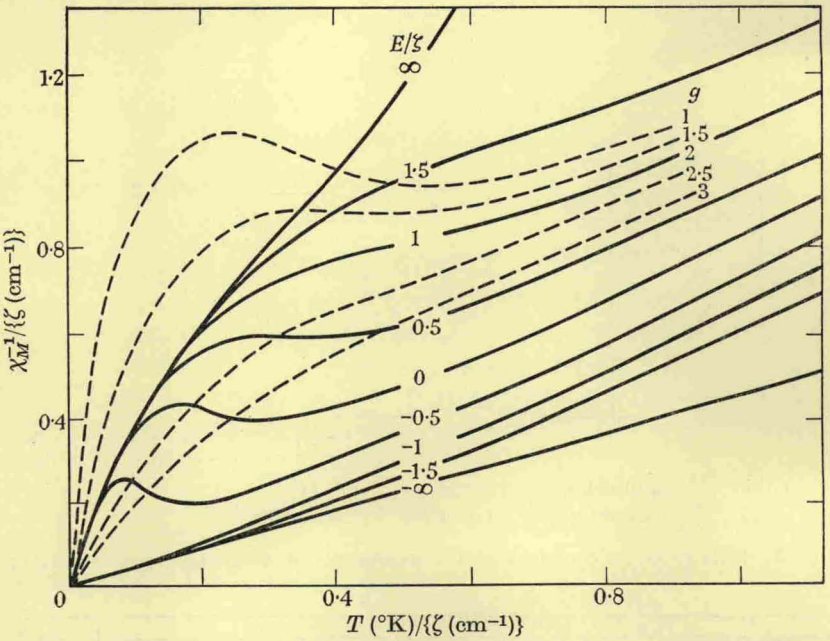

FIGURE 5. Calculated values of χ_M^{-1}. Full lines: $g = 2$ with various values of E/ζ. Broken lines: $E/\zeta = 1$ with various values of g.

figure 4 may produce maxima and minima in curves of $\chi_M(T)$ and $\chi_M^{-1}(T)$. Such maxima and minima are of obvious diagnostic value, and they are predicted in the present instance when $|E|/\zeta < 1$.

The generality of this last inequality requires some qualification. The parameter g was introduced into figure 3 and equation (1) as one of the many possible ways of distorting a doubtless idealized model. The broken curves in figure 5, in which we have assumed a constant value of E/ζ, namely $+1$, and various values of g, shows that the maximum and minimum in χ_M^{-1} may develop, for $|E|/\zeta > 1$, if g differs sufficiently from 2. On the other hand, g-values differing from 2 by more than 0·5 for a low-spin octahedral complex would be unexpected. We conclude that an experimental observation of a maximum and minimum in $\chi_M^{-1}(T)$ is a sensitive test for nearly coincident $^2T_2$ and $^6A_1$ states. At the same time, the precise details of the model assumed in figure 3 are seen not to be critical. Indeed, calculations show that maximum-minimum behaviour could be produced in this system even if spin-orbit interaction were neglected entirely, provided one admitted a sufficiently low value