APPENDIX III

Calculation of Magnetic Susceptibility

Calculation of magnetic susceptibility reported in Sec. 11 was accomplished by a computer program SUSP. The algorithm closely follows Griffith's derivation (Section 10-1 of G61) of Van Vleck formula (Eq. 9.7):

\[
\chi_\alpha = N \frac{\sum \left\{ \left( \frac{E_i^2}{kT} - 2E_i^2 \right) e^{-E_i^2/kT} \right\} \epsilon}{\sum \epsilon e^{-E_i^2/kT}}
\]

where \( \alpha = x, y, z \). \( E_i^2 \) and \( E_i^2 \) are also known low frequency and high frequency terms. These are calculated as follows:

(a) First using a convenient basis set (in our case strong field basis set), the Hamiltonian matrix \( H \) and magnetic moment operator matrix \( \lambda_{\alpha} = \beta (kL_\alpha + S_\alpha) \) are evaluated.

(b) \( H \) is a diagonalized. Its eigenvalues give \( E_i^2 \). Using the matrix of eigenvectors of \( H, U \), the magnetic moment operator matrix \( G \) is transformed to the new bases set, \( G' = U^TGU \)

(c) The portion of \( G' \) corresponding to degenerate eigenvalues \( E_i^6 \) is then diagonalized. Let \( V \) be the matrix of eigenvectors which diagonalises the degenerate block of \( G' \).

\[
G' = V^T G V
\]
After these steps, the magnetic moment operator matrix will have the form:

\[
\begin{pmatrix}
\eta_1 & 0 & \cdots & 0 & a_{1n} & a_{1n+1} & \cdots & a_{1m} \\
0 & \eta_2 & \cdots & 0 & a_{2n} & a_{2n+1} & \cdots & a_{2m} \\
\vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\
0 & 0 & \cdots & \eta_n & a_{nn} & a_{n+1} & \cdots & a_{nm}
\end{pmatrix}
\]

The diagonal elements \( \eta_i \) give the low frequency terms \( E_i^1 \), and the corresponding row elements \( a_{ij} \) give high frequency terms.

\[
E_i^2 = \sum_j \left| \frac{a_{ij}}{E_j^0 - E_i^0} \right|^2
\]

Weighting with the Boltzmann factor and summing one gets the susceptibility.