Fundamentals of Magnetism
WS 24/25, Uni Leipzig

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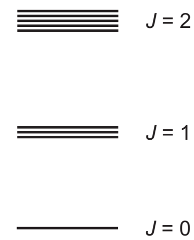
Problem sheet 3: Van Vleck paramagnetism, Pauli paramagnetism **24.11.2024**

You can use both the SI or CGS units, but you may find CGS easier whenever magnetic moments and magnetic susceptibilities are involved.

3.1. $J = 0$ magnetism (8 P)

In its low-spin state, the octahedrally coordinated Ru^{4+} ($3d^4$) features $S = 1$ and $L = 1$, giving rise to three multiplets with $J = 0, 1, 2$, as shown in the figure (note that $J = 0$ has the lowest energy, at odds with the third Hund's rule; this is because we calculate L within the t_{2g} shell where the third Hund's rule is "reversed"). The splitting between these multiplets is due to the spin-orbit coupling, $\lambda\,\hat{\mathbf{L}}\,\hat{\mathbf{S}}$.



(a) Calculate the expectation values of $\hat{\mathbf{L}}\,\hat{\mathbf{S}}$ and determine the energies of the $J = 1$ and $J = 2$ multiplets.

(b) Use the Clebsch-Gordan coefficients to decompose the states of the J-multiplets into the $|l_z, s_z\rangle$ states.

(c) Calculate the matrix elements $\langle 0|g_L\hat{L}_z + g_S\hat{S}_z|n\rangle$ where $|0\rangle$ is the ground state ($J = 0$) and $|n\rangle$ are the excited states of the $J = 1$ and $J = 2$ multiplets. Note that you have to use $g_S = 2$ but $g_L = -1$, the latter is again a consequence of considering only the t_{2g} shell and not the spherically symmetric atom.

(d) Using $\lambda = 0.1$ eV, determine the Van Vleck susceptibility of Ru^{4+}. Compare the result to the experimental data for K_2RuCl_6 from Fig. 1 of Phys. Rev. Lett. 127, 227201 (2021).

(e) Try to get the temperature dependence by including the magnetism of the excited states.

3.2. Pauli paramagnetism, also at elevated temperatures (7 P)

Use the free-electron model to analyze Pauli paramagnetism of sodium ($a = 4.225$ Å, bcc lattice = 2 atoms per unit cell, $Z = 1$ valence electron per atom).

(a) Calculate the density of states at the Fermi level, $N(\varepsilon_F)$, and the corresponding Pauli susceptibility at 0 K.

(b) Obtain the lowest-order correction to this result at finite temperatures. To this end, use the expression from the lecture,

$$M = \mu_B^2\, B \times \int N'(\varepsilon) f(\varepsilon)\, d\varepsilon$$

and the Sommerfeld expansion,

$$\int_0^\infty H(\varepsilon) f(\varepsilon)\, d\varepsilon = \int_0^{\varepsilon_F} H(\varepsilon)\, d\varepsilon + \frac{\pi^2}{6}(k_BT)^2 H'(\varepsilon_F)$$

where $H(\varepsilon)$ is an arbitrary function and $f(\varepsilon)$ is the Fermi-Dirac distribution.

(c) Determine the correction to the magnetic susceptibility of Na at 300 K.

The electron concentration, Fermi energy, and density of states can be determined using the lattice parameter of sodium. Check any solid-state physics textbook if you are not sure (or forgot) how to do this.

3.3. Stoner enhancement (5 P)

Consider the Stoner's model from lecture 4 and augment its energy with an additional term due to magnetic field,

$$\Delta E = \frac{1}{2}N(\varepsilon_F)(\delta E)^2(1 - UN(\varepsilon_F)) - MB$$

where $M = \mu_B(n_\uparrow - n_\downarrow) = \mu_B N(\varepsilon_F)\,\delta E$.

(a) Minimize ΔE with respect to M and show that the magnetic susceptibility is given by

$$\chi = \frac{\chi_\mathrm{P}}{1 - UN(\varepsilon_F)}$$

where χ_P is the Pauli susceptibility. The correction to χ_P is the Stoner enhancement that takes place when electron-electron interactions are present, yet not strong enough to induce the ferromagnetic instability in a metal.

(b) Use the experimental magnetic susceptibility of palladium, $\chi = 5.4 \times 10^{-4}\,\mathrm{cm}^3/\mathrm{mol}$, to determine U, the energy of the electron-electron repulsion. Express the result in eV. Use $N(\varepsilon_F) = 2\,\mathrm{eV}^{-1}$/atom, the density of states at the Fermi level.