

Open Systems

Spins, Spin Lattice Relaxation, and Non-Radiative Transitions

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This Lecture will be concerned with how to handle real systems. There will be very little (if any) new quantum mechanics, but there will be plenty of challenges, even without resorting to the computer. Most of the issues are discussed in these sources (where full references are given), and I shall not reproduce the formal analysis.

1. A M Stoneham 1975 (1985, 2001) *Theory of Defects in Solids* Oxford: Oxford University Press. *Most of the relevant material is in Chapter 14*
2. A M Stoneham *Non-Radiative Transitions in Semiconductors* 1981 Rep. Prog. Phys. 44, 1251-1295. *This updates parts of ref 1 and gives more detail than ref 3.*
3. N Itoh and A M Stoneham 2000 *Materials Modification by Electronic Excitation* Cambridge: Cambridge University Press *Most of the relevant material is in Chapter 3*

The Power Point file is intended to save you having to try to copy complex figures.

Ideas of spin

Isolated spin and spin analogues: interactions with an electromagnetic field

Spins interacting with each other

Spin or other local excitations interacting with lattice vibrations

The Spin Hamiltonian 1

Basic Idea 1: If there are N states with very low energy, then we can write an effective Hamiltonian for N basis states, with the effects of the distant states included by perturbation theory.

Basic Idea 2: We can write this effective Hamiltonian in terms of spin operators with *effective* spin S such that $N = 2S+1$.

Note S does not need to be the actual spin of the system.

The Spin Hamiltonian 2: Why bother?

1. Spin matrices are well-known and easy to manipulate
2. The spin Hamiltonian puts everything into a format to make comparison of experiment and theory easy.
 - It can be used to predict transition energies and probabilities so experimenters can get a full empirical fit.
 - The key parameters of the fit are what the theorists try to predict

The Spin Hamiltonian 3: Some complications

Usually the spin Hamiltonian relates to a single defect or spin

Equations are often written for an ensemble of spins

How do we handle the interactions?

- Is it true that the magnetic dipoles deduced from Zeeman energies

are the same as the dipoles describing the interactions between spins?
 - The full dipole-dipole interaction includes $S_z S_z$, $S_+ S_-$, $S_- S_+$ and similar terms. What do they do

Rate equations for a two-level system

$$dN_L/dt = W_{UL}N_U - W_{LU}N_L$$

$$dN_U/dt = W_{LU}N_L - W_{UL}N_U$$

$$\text{Let } N = N_L - N_U$$

Subtract one equation from the other

$$dN/dt = - (W_{UL} + W_{LU}) N = -N/\tau$$

$$\text{Relaxation rate } 1/\tau = (W_{UL} + W_{LU})$$

Rate equations and better

Only *occupancies* N_U and N_L involved: phases and wavefunction information hidden or lost.

What interaction causes the transitions
 Where does the energy go?

To which states do $|L\rangle$ and $|U\rangle$ correspond? Can we really separate them, the energy sink, and the interaction?
 And can we *predict* $1/\tau$ as a number?

Density matrix versions

Since we're talking about a two-level system, let's use \mathbf{S} as the ensemble average of the spin for spins $1/2$ interacting with a heat bath and a magnetic field. The density matrix has the form

$$\rho = \frac{1}{2} \begin{bmatrix} 1 + S_z & S_x - i S_y \\ S_x + i S_y & 1 - S_z \end{bmatrix}$$

and the equivalent equation of motion is

$$-i\hbar \partial\rho/\partial t = [H_z, \rho] - i\hbar/\tau [\rho - \rho_0]$$

where there's a static field and an oscillating field

$$H_z = \hbar\omega_0 S_z + \frac{1}{2} \hbar\omega_1 [S_+ \exp(i\omega t) + S_- \exp(-i\omega t)]$$

Solve for energy absorption from the field

The energy absorption can be calculated from $\mathbf{S} \cdot d\mathbf{H}/dt$
 In the steady state, the energy absorption is proportional to

$$[\omega_1^2 \omega_0 \omega \tau] / [1 + \omega_1^2 \tau^2 + \tau^2(\omega - \omega_0)^2]$$

which leads to a maximum energy absorption at frequency

$$\omega_m = 1/\tau \sqrt{1 + (\omega_1^2 + \omega_0^2)\tau^2}$$

Resonant absorption: for $1 \gg \tau \sqrt{(\omega_1^2 + \omega_o^2)}$
 $\omega_m = \sqrt{(\omega_1^2 + \omega_o^2)}$ or, if $\omega_1 \ll \omega_o$ (usually true) $\omega_m = \omega_o$
Non-resonant absorption for $1 \gg \tau \sqrt{(\omega_1^2 + \omega_o^2)}$
 $\omega_m = 1/\tau$

Non-radiative Transitions 1 Types of transition

Normal modes and Reaction Coordinates
Configuration coordinate diagrams
Cooling transitions: one energy surface
Two energy surfaces
Strong and weak coupling. Choice of Hamiltonian
Energy level structures in relation to phonons
Spin-lattice relaxation

Non-radiative transitions 2 Simple arguments

Branching ratios
Promoting and accepting modes
Lineshape function and the absorption of energy from the sudden switch-on of a force
Bounds on absolute rates