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Autor: Vojta, Günter
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Overhauser Nuclear Polarization Effect and Spin Temperatures

by Günter VOJTA

Leipzig

Barker et d'autres chercheurs ont réussi à traiter l'effet Overhauser par la méthode des potentiels chimiques. Nous montrons qu'on obtient simplement les résultats principaux concernant l'effet Overhauser et le principe de production minimum d'entropie avec l'hypothèse de la température de spin.

In interesting papers Barker and others [1] express doubts to the validity of spin temperatures and give a very simple and elegant proof of the Overhauser effect using the notion of chemical potentials. In opposition to them we will achieve here some considerations in favour of the notion of spin temperature.

One of the well-known objections against this concept is the following: For the Overhauser effect is essential a strong coupling between electron and nuclear spins, and therefore an increase of the electron spin temperature T_e should be followed by an increase of the nuclear spin temperature T_n , in contrary to the observed lowering. But this argument is not correct. The coupling between the two spin systems is not a thermal contact in the sense of thermodynamics, for *both* spins deliver energy (to the lattice) or receive energy during an elementary process, i.e. simultaneous flips of electron and nuclear spin. Certainly the introduction of spin temperatures requires further discussion here, because the total system is in a (stationary) nonequilibrium state [2, 3].

Till now there are only few thorough papers on (statistical) thermodynamics of spin systems using spin temperatures. A very simple derivation of the Overhauser effect on this basis was given by Van Vleck [4]. Perhaps just as simple is the following derivation, for which only the conservation theorem of the z -component of the total (electron + nuclear) spin is necessary; this theorem is valid for the Overhauser interaction

mechanism. It is sufficient to deal with the simplest (hypothetical) case: nuclei with spin $\frac{1}{2}$ and electrons obeying Boltzmann statistics; nuclei with spin I and use of Fermi statistics do not cause essential change in the proof. Let N_1 and N_2 be the numbers of nuclear spins with $I_z = +\frac{1}{2}$ resp. $-\frac{1}{2}$ and N_+ and N_- the numbers of electron spins with $S_z = +\frac{1}{2}$ resp. $-\frac{1}{2}$; $N_1 + N_2 = N_+ + N_- = N$. The corresponding energy niveaus are

$$-\frac{1}{2} \gamma_n \hbar H_0, \quad +\frac{1}{2} \gamma_n \hbar H_0,$$

and

$$-\frac{1}{2} \gamma_e \hbar H_0, \quad +\frac{1}{2} \gamma_e \hbar H_0$$

(γ_n, γ_e : gyromagnetic ratios of the nuclei resp. the electrons, $\gamma_e < 0$!). Then we have $N_1/N_2 = \exp 2\delta_{on}$, where $\delta_{on} = \gamma_n \hbar H/2kT_n$.

It follows

$$N_1 = N/[\exp(-2\delta_{on}) + 1], \quad N_2 = N/[\exp(2\delta_{on}) + 1] \quad (1)$$

and correspondingly N_+ and N_- . The conservation law

$$\frac{1}{2} N_1 - \frac{1}{2} N_2 + \frac{1}{2} N_+ - \frac{1}{2} N_- = \text{const} \quad (2)$$

is valid for the relaxation process. A strong microwave field may be switched on for a very short time, by which T_e is raised momentarily. After that the relaxation sets in: Lowering of T_e down to the lattice temperature T_l and of T_n starting from the value T_l . From (2) we find with the help of (1) and with $(2\delta_{on})^2 \ll 1$, $(2\delta_{on})^2 \ll 1$:

$$\frac{|\gamma_e|}{T_e(t)} - \frac{\gamma_n}{T_n(t)} = \text{const},$$

valid for every time t . If we let $T_e =$ electron spin temperature at the beginning and $T_n =$ nuclear spin temperature at the end of the relaxation process, it follows

$$\frac{|\gamma_e|}{T_e} - \frac{\gamma_n}{T_l} = \frac{|\gamma_e|}{T_l} - \frac{\gamma_n}{T_n}.$$

In the stationary state $T_e(t)$ is fixed on the value T_e , but $T_n(t)$ naturally has the value T_n . So we have

$$\frac{1}{T_n} = \frac{\gamma_n + |\gamma_e|}{\gamma_n T_l} - \frac{|\gamma_e|}{\gamma_n T_e}.$$

This is equ. (8) of Van Vleck [4].

In a just as simple manner we can show the (approximate) validity of the principle of minimum entropy production [3, 5]. So it is evident, that with the concept of spin temperature we can get essential results in a simple and, in our opinion, correct way. The considered problems shall be treated in detail in a later paper.

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