Physica B 165&166 (1990) 231-232
North-Holland

CROSS-RELAXATION IN PARAMAGNETIC CRYSTALS AT LOW TEMPERATURES

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The well- known phenomena of cross-relaxation between two spin-systems in paramagnetic crystals is theoretically investigated at low temperatures when the Zeeman energy of spins exceeds their heat energy, but there is no order due to spin-spin interactions in spin system.

1. INTRODUCTION

The cross-relaxation (CR) takes place in paramagnetic crystals containing two kinds of spins (S spins and I spins) with close resonant frequencies ω_s and ω_i. This phenomena consists in an energy exchange between the Zeeman subsystems and the reservoir of spin-spin interactions (SSI). The kinetics of spin system is determined by its thermodynamic properties only.

In the high temperature (HT) approximation ($\hbar\omega_m \ll kT_0$, where $m=s,i$, T_0 is a lattice temperature) the theory of CR was developed in (1). Accordingly to this theory the Zeeman subsystems and the reservoir of SSI are statistically independent. A coupling between subsystems appears in the result of CR.

At low temperatures (LT) ($\hbar\omega_m > kT_0$) the subsystems become thermodynamically dependent (2,3). The strong thermodynamic coupling causes that any change of one subsystem state must be accompanied by changes of states of others. So we can expect the essential increase of CR efficiency.

2. THERMODYNAMICS OF SPIN SYSTEM AT LT

Let us consider a concentrated paramagnetic crystal containing S and I spins (S=I=1/2). The Hamiltonian of problem is:

$$H=H_s+H_i+H_{ss},\quad H_s=\omega_s\sum_j S_j^z,\quad H_i=\omega_i\sum_\alpha I_\alpha^z$$

$$H_{ss}=\tfrac{1}{2}\sum_{ij}(A_{ij}S_i^zS_j^z+B_{ij}S_i^+S_j^-)+$$

$$+\tfrac{1}{2}\sum_{\alpha\beta}(A_{\alpha\beta}I_\alpha^zI_\beta^z+B_{\alpha\beta}I_\alpha^+I_\beta^-)+\sum_{i\alpha}A_{i\alpha}S_i^zI_\alpha^z$$

The SSI constants include the exchange and the dipole-dipole interactions between spins.

If a lattice temperature is equal to zero then the magnetizations of spin sublattices

$$p_s=-2\langle S_j^z\rangle \text{ and } p_i=-2\langle I_\alpha^z\rangle$$

are full. At low temperatures magnetizations are about full and number of spins of every kind turned over a direction of magnetic field is small. That is why we can speak about elementary excitations, which can be named by spin excitons(4). It is necessary to note that such excitations result from the Zeeman interaction with magnetic field by contrast to magnons existing due to SSI. Thus we have two non-interacting types of spin excitons.

Any non-equilibrium state of spin system can be considered as the state with some number of non-equilibrium excitons of every type. In the quasi-equilibrium density operator we can introduce the chemical potentials μ_s and μ_i (2,3) describing a number of flipped spins:

$$\rho_q = Q_q^{-1}\exp(-\beta(H-\mu_s\textstyle\sum_j S_j^z-\mu_i\sum_\alpha I_\alpha^z))$$

where β is an inverse spin temperature. Such form of quasi-equilibrium distribution takes place due to the fact of the availibility of three first integrals. A physical mechanism establishing this quasi-equilibrium is a spin rotation in local fields like the HT theory. The only characteristic feature concludes in an appearence of the molecular field at LT, which causes the ordering of local fields.

In the regular paramagnetic crystal a wave vector is considered to be a good quantum number and S-excitons and I-excitons are characterized by wave vectors $\mathbf{k}_s$ and $\mathbf{k}_i$ respectively. The energy spectrum of each type can be found in the molecular field approximation. The obtained expressions describe two exciton zones. For each zone we can find out the average