LOW-FREQUENCY RESPONSE OF THE SINGLET-TRIPLET SYSTEM IN THE PARAMAGNETIC PHASE

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The expressions for the longitudinal dynamic magnetic susceptibility of the singlet-triplet system in the paramagnetic phase are obtained within the linear response theory by introducting two different single-ion relaxation times which appear due to the interaction between the magnetic system and a thermostat. The dynamical properties of the system can be interpreted in terms of the Van Vleck mode scattered on Curie – Langevin fluctuation i.e. a fluctuation of the diagonal part of the magnetic moment operator $\hat{J}^z$. In the vicinity of the Curie point and in a low-frequency region we obtain the following results: (*i*) the central peak with the lifetime increasing to infinity at T_c; (*ii*) the modified Van Vleck mode with frequency decreasing to zero as $T \to T_c$, but with finite damping. This mode passes into a central peak at a temperature $T_0 \geqslant T_c$ defined as that at which the real part of this mode vanishes.

1. *Introduction*

The aim of this paper is to describe the influence of the single ion relaxation time τ_2 on the magnetic excitation spectrum in the paramagnetic phase of the singlet-triplet systems. Time τ_2 is related to transitions of the operator within the triplet states. The above mentioned problem was partly presented in our previous paper [1]. Spectral intensity obtained by the method presented in [1, 2] is studied. The expression for the longitudinal dynamical susceptibility is obtained within the linear response theory by introducting two different single ion relaxation times (τ_1, τ_2), which appear due to the interaction between the magnetic system and the thermostat.

The susceptibility is directly related to the spectral intensity via the fluctuation-dissipation-theorem. On the basis of the general spectral intensity formula (see: [3, 4]) and the results of paper [1] one can write:

$$S^+(q, \omega) = \pi^{-1} \coth \frac{\omega}{2kT} \operatorname{Im} \chi(q, \omega), \tag{1}$$

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where S^{+} is the spectral intensity and Im $\chi(\boldsymbol{q}, \omega)$ — the imaginary part of the magnetic susceptibility, which is expressed by the following formula:

$$\chi(\boldsymbol{q}, \omega) = \frac{\chi_{VV}(\omega)+\chi_{C-L}(\omega)}{1-2J(\boldsymbol{q})\,(\chi_{VV}(\omega)+\chi_{C-L}(\omega))}, \tag{2}$$

where

$$\chi_{VV}(\omega) = \frac{\chi_{VV}\Delta^2}{\Delta^2-\omega^2+2i\omega\gamma}. \tag{2a}$$

$\chi_{VV}(\omega)$ is the Van Vleck single ion dynamic suspcetibility related to the singlet-triplet transition of the $\hat{J}^z$ operator (i.e. inelastic term of the response) and χ_{VV} is the single ion static isothermal Van Vleck susceptibility. $\gamma = 1/\tau_1$ and τ_1 is the relaxation time for transitions between the singlet and the triplet. Δ is the energy difference between the triplet and the singlet energy levels.

$$\chi_{C-L}(\omega) = \frac{\chi_{C-L}}{1+i\omega\tau_2} \tag{2b}$$

$\chi_{C-L}(\omega)$ is the Curie-Langevin susceptibility, which is related to the intratriplet transitions (i.e. the elastic term of the response) and τ_2 is the relaxation time determining the dynamical scale of the transitions due to the interaction with the thermostat. χ_{C-L} is the static Curie-Langevin susceptibility, $J(\boldsymbol{q})$ — the Fourier transform of the exchange integral.

As it was shown γ is the damping parameter for the Van Vleck mode. If this parameter is small enough then its value will not influence the qualitative results obtained below and therefore we put γ equal to zero ($\gamma = 0$).

We also assume that the system is ferromagnetic below the transition temperature which is defined as the temperature at which the static isothermal susceptibility diverges for $\boldsymbol{q} = 0$ i.e.:

$$1-2J(0)\,(\chi_{VV}+\chi_{C-L}) = 0. \tag{3}$$

Now, we can write the following formula for the spectral intensity:

$$S^{+}(\boldsymbol{q}, \omega) = \frac{\pi^{-1}\coth\dfrac{\omega}{2kT}\,\omega\tau_{2q}\left\{\chi_{VV}\Delta^2(\bar{\omega}_0^2-\omega^2)+\dfrac{\omega_0^2-\omega^2[\omega^2\chi_{C-L}-\Delta^2(\chi_{VV}+\chi_{C-L})]}{(1-1J(\boldsymbol{q})\chi_{C-L})}\right\}}{(\bar{\omega}_0^2-\omega^2)^2+\omega^2\tau_{2q}^2(\omega_0^2-\omega^2)^2}$$

where

$$\tau_{2q} = \frac{\tau_2}{1-2J(\boldsymbol{q})\chi_{C-L}}, \tag{4a}$$

$$\bar{\omega}_0 = \sqrt{\omega_0^2-L}, \tag{4b}$$

$$L = 4J^2(\boldsymbol{q})\Delta^2\chi_{VV}\frac{\chi_{C-L}}{1-2J(\boldsymbol{q})\chi_{C-L}}, \tag{4c}$$

$$\omega_0 = \Delta\sqrt{1-2J(\boldsymbol{q})\chi_{VV}}\,. \tag{4d}$$

$S^{+}(\boldsymbol{q}, \omega)$ is analyzed numerically in detail with no initial simplifications nor assumptions.

2. *Numerical results and their discussion*

In our model three parameters of the system i.e.: Δ, $V = \frac{J(0)}{\Delta}$ and τ_2 are introduced.

The spectral intensity is calculated for a singlet-triplet system (P_r^{3+}; $J = 4$ in cubic environment) in a wide range of the above mentioned parameters. We find that τ_2 plays a very important role because it can change completely the magnetic excitation spectrum in the low-frequency region. As has been already mentioned τ_2 is the relaxation time related to the intratriplet transitions (see: Eq. (2b)) i.e. with fluctuation of the diagonal part intra-triplet part of the magnetic moment operator $\hat{J}^z$ expressed in $g_J\mu_B$ units.

We obtain the following results:

1. $\omega\tau_2 \gg 1$ and $\omega_0\tau_{2q} \gg 1$.

Under this condition appears the Van Vleck mode only (see: Eq. (4d)). Fig. 1 shows the Van Vleck mode energy as a function of temperature. Approaching T_c from above one

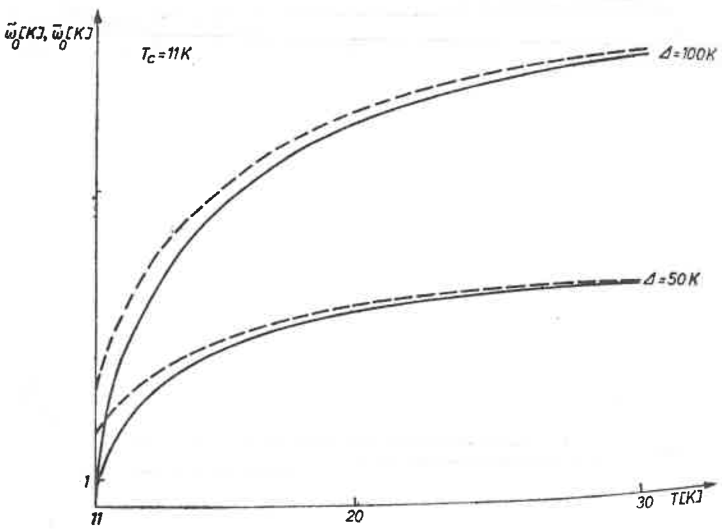


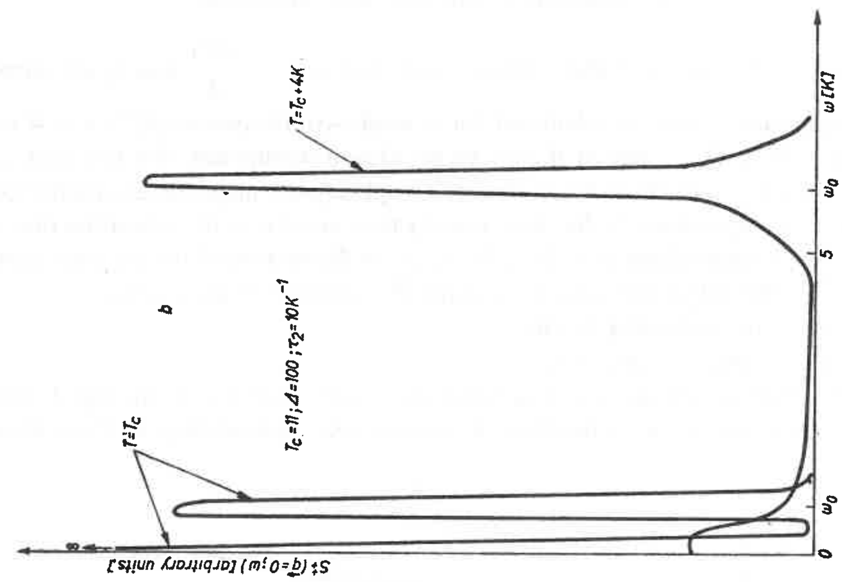
Fig. 1. Temperature dependence of the Van Vleck mode (ω_0) and soft mode ($\bar{\omega}_0$) energies for several values of Δ. Dashed lines — energies of the Van Vleck mode. Solid lines — energies of the soft mode. $T_c = 11$ K; $\Delta = 50$ K, $\Delta = 100$ K

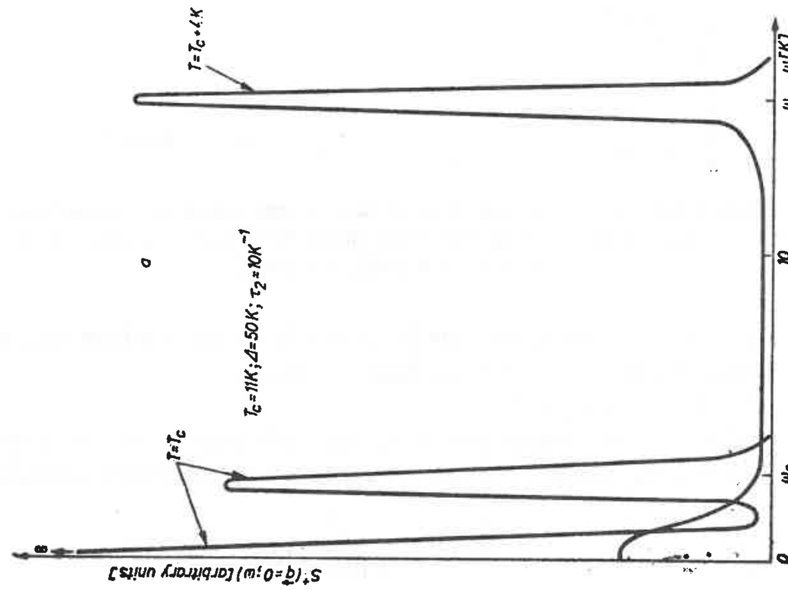
notices that the Van Vleck mode does not become soft. In the low-frequency region i.e. $\omega\tau_2 \ll 1$ we can arrive at one of the two following cases:

2. $\omega\tau_2 \ll 1$ but $\omega_0\tau_{2q} \gg 1$

then the spectral intesity is dominated by a strong elastic peak at $\omega = 0$ (central peak). Spectral intensity for the central peak can be expressed by the Debye formula:

$$S^+(q, \omega) \sim \frac{\tau^*}{1+\omega^2\tau^{*2}}, \tag{5}$$


T=Tc+4K
ω [K]
ω0
5
Tc=11; Δ=100; τ2=10K^-1
b
T=Tc
ω0
0
S↑(q=0;ω) [arbitrary units]
∞


T=Tc+4K
ω [K]
ω0
10
Tc=11K; Δ=50K; τ2=10K^-1
a
T=Tc
ω0
0
S↑(q=0;ω) [arbitrary units]
∞

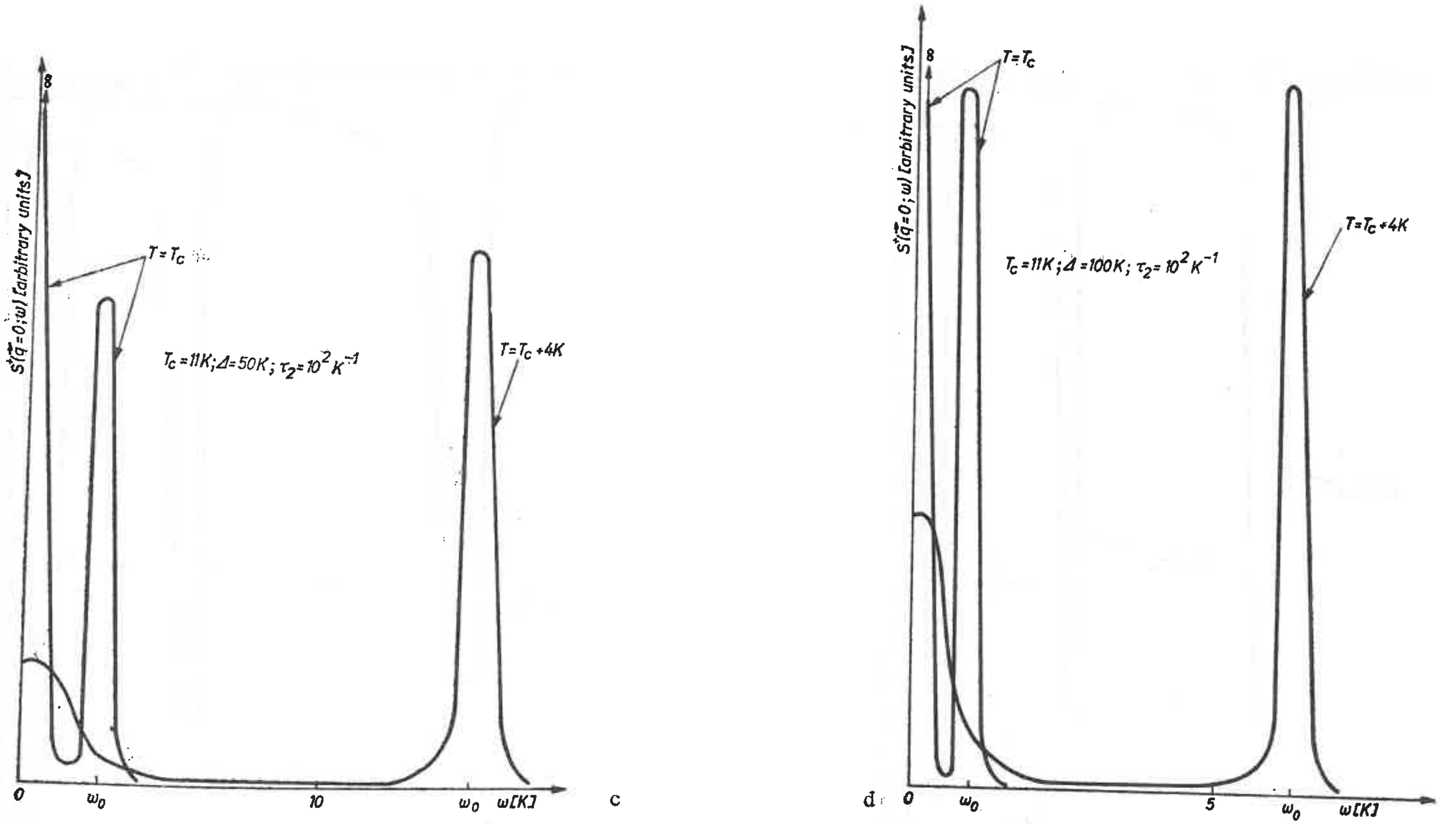
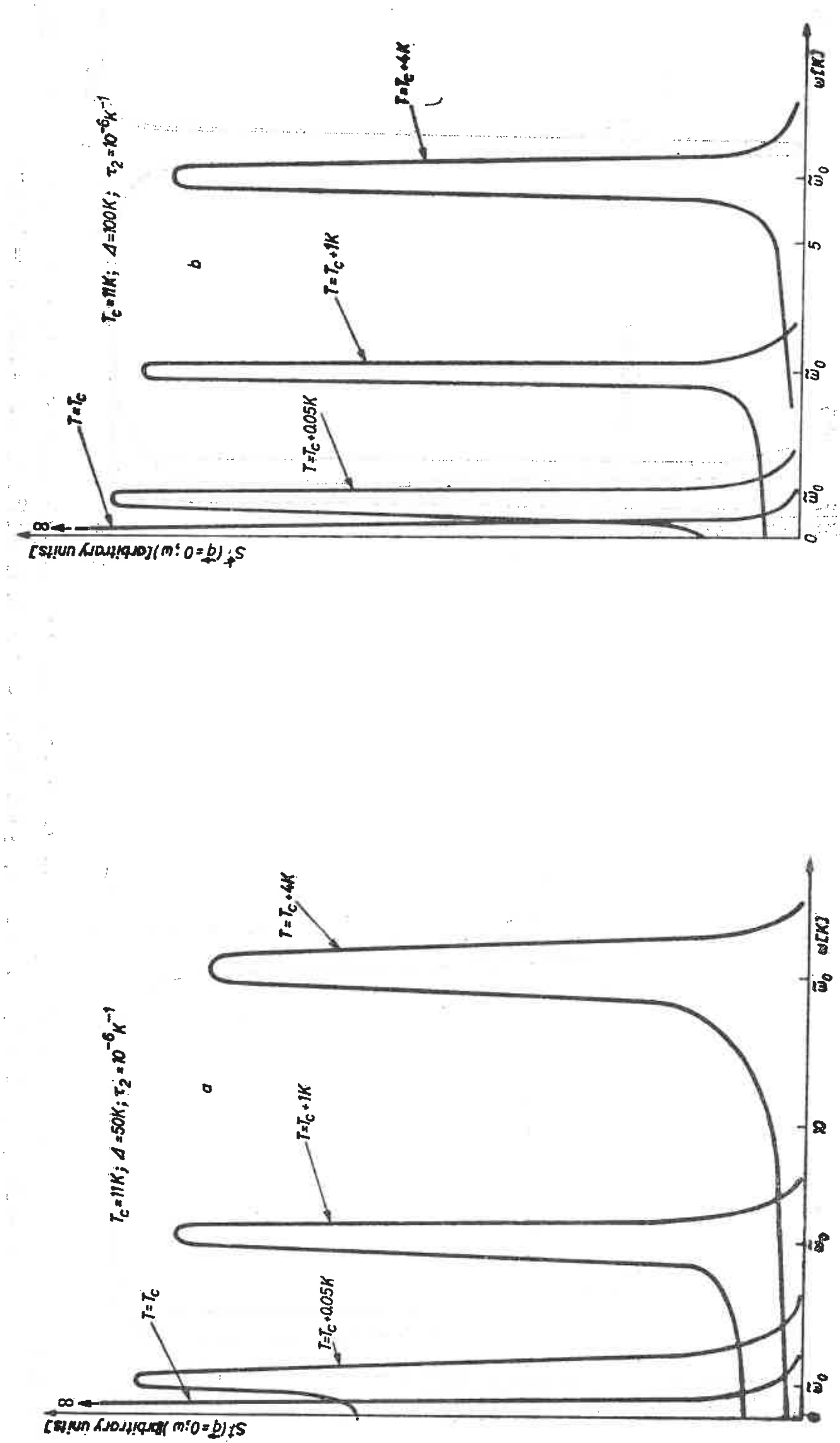


Fig. 2. The spectral intensities for the central peak behaviour plotted as functions of frequency for several values of Δ, τ_2 and temperature. $T_c = 11$K; a) $\Delta = 50$ K, $\tau_2 = 10$ K^{-1}, b) $\Delta = 100$ K, $\tau_2 = 10$ K^{-1}, c) $\Delta = 50$ K, $\tau_2 = 10^2$ K^{-1}, d) $\Delta = 100$ K, $\tau_2 = 10^2$ K^{-1}


S(q=0;ω)[arbitrary units]
∞
T=T_c
T=T_c+0.05K
T=T_c+1K
T=T_c+4K
T_c=11K; Δ=100K; τ_2=10^-6 K^-1
b
0
5
ω̃_0
ω[K]
T_c=11K; Δ=50K; τ_2=10^-6 K^-1
a
10

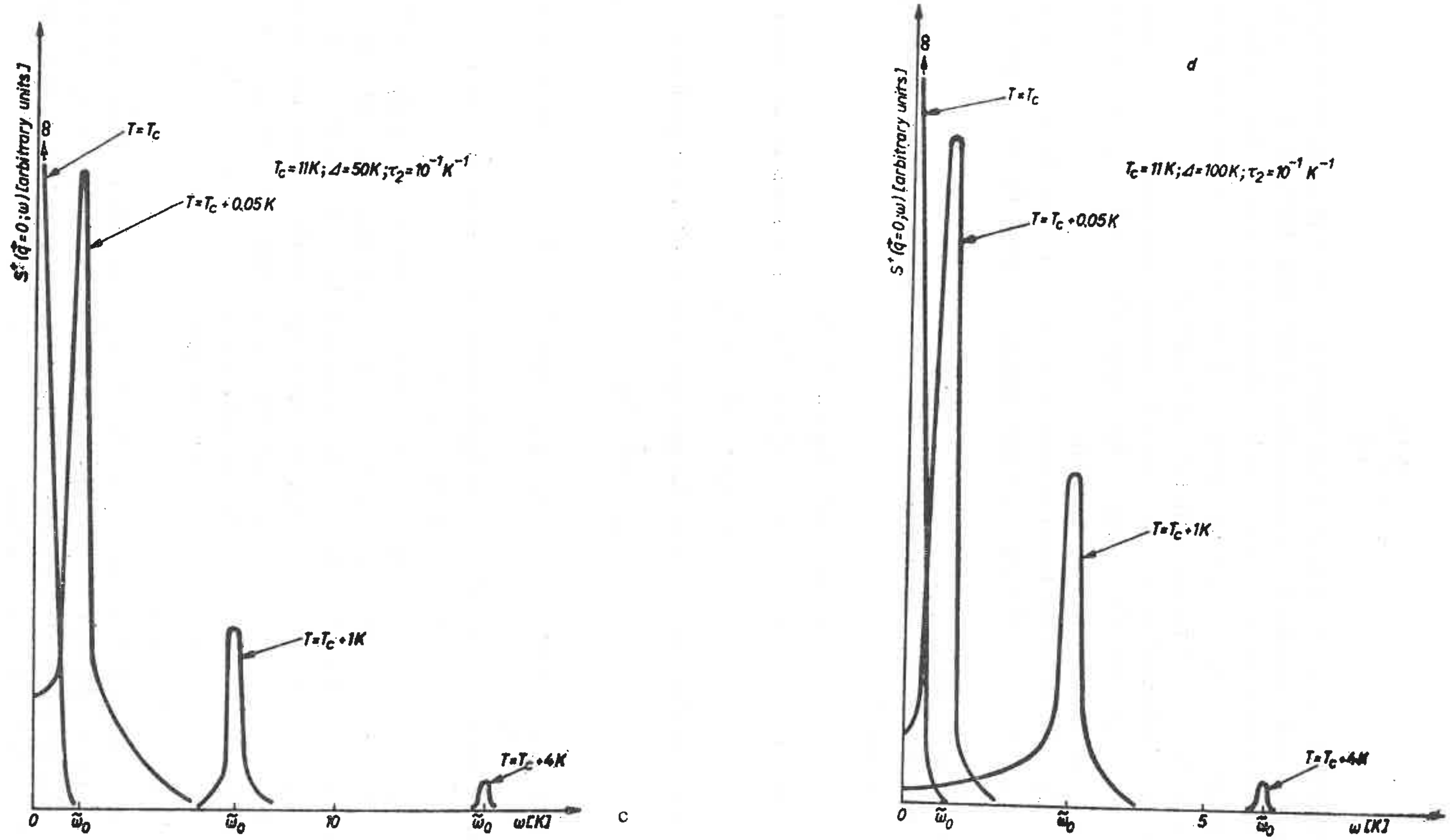


Fig. 3. The spectral intensities for the soft mode behaviour plotted as functions of frequency for several values of Δ, τ_2 and temperature. $T_c = 11$ K; a) $\Delta = 50$ K, $\tau_2 = 10^{-6}$ K^{-1}, b) $\Delta = 100$ K, $\tau_2 = 10^{-6}$ K^{-1}, c) $\Delta = 50$ K, $\tau_2 = 10^{-1}$ K^{-1}, d) $\Delta = 100$ K, $\tau_2 = 10^{-1}$ K^{-1}

with

$$\tau^* = \frac{\omega_0^2 \tau_{2q}}{\bar{\omega}_0^2}, \tag{5a}$$

τ^* is the modified relaxation time. Eq. (5) shows clearly the central peak i.e. zero-frequency fluctuations at the static stability limit. The width of this peak decreases to zero and its height grows to infinity at the Curie point. This is illustrated in Fig. 2. We find that the central peak behaviour is due to the Curie-Langevin fluctuation i.e. the fluctuation of the diagonal contribution to the magnetic moment operator $\hat{J}^z$.

A lifetime of this fluctuation becomes very large as the Curie temperature is reached and at T_c it tends to infinity. Similar results have been obtained by Egami and Brooks (see: [5]) in the mode-mode coupling approximation.

The total magnetic excitation spectrum consist of the Van Vleck mode in the high--frequency region and the central peak in the low-frequency region. The results are shown in Fig. 2.

3. $\omega\tau_2 \ll 1$ and $\bar{\omega}_0 \tau_{2q} \ll 1$.

The normal mode energies are defined as the poles of $\chi(\boldsymbol{q}, \omega)$. The denominator of Eq. (2) must be equal to zero i.e.:

$$\bar{\omega}_0^2 - \omega^2 - i\omega\tau_{2q}(\omega_0^2 - \omega^2) = 0. \tag{6}$$

Under the conditions (3) we obtain from Eq. (6) the following normal-mode frequencies

$$\omega_q^{1,2} = \sqrt{\bar{\omega}_0^2 - \gamma^2(T)} - i\gamma(T), \tag{7}$$

where $\gamma(T)$ is the damping parameter of the modified Van Vleck mode ($\bar{\omega}_0$) and

$$\gamma(T) = \frac{\omega_0^2 - \tau_{2q}}{2}. \tag{7a}$$

The temperature variation of $\bar{\omega}_0$ and ω_0 are shown in Fig. 1. The temperature variation of τ_{2q} is shown in Ref. [1].

As it follows from Eq. (7a) $\gamma(T)$ is very slightly temperature-dependent and it remains finite at T_c i.e. the behaviour of $\gamma(T)$ is noncritical.

From Eq. (7) it is seen that $S^+(q = 0, \omega)$ shows two peaks at $\omega = \sqrt{\bar{\omega}_0^2 - \gamma^2(T)}$ but there is always the temperature $T_0 \gtrsim T_c$ defined as the one at which the real part of the normal mode energy vanishes (see: Eq. (6)). In the range between T_0 and T_c temperatures the relaxation type response is obtained i.e. the zero-frequency fluctuations central peak.

The "pure" soft mode behaviour can only appear in the system if $\gamma(T)$ equals to zero i.e. τ_{2q} is equal to zero and then $T_0 = T_c$. The behaviour of S^+ $(q = 0, \omega)$ is illustrated in Fig. 3. When $T \to T_c$ the peak in S^+ $(q = 0, \omega)$ shifts towards $\omega = 0$ and the peak grows to infinity with its width decreasing to zero.

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