acta physica slovaca vol. 44 No. 4/5, 269 – 274 August 1994

RESPONSE OF QUANTUM SYSTEMS WITH INCOMMENSURABLE MODULATED PHASES[1]

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Received 27 April 1994, accepted 2 June 1994

Theory of response of quantum systems, ordered in incommensurate phases, at low temperatures and low frequencies was developed. The observed linear dependence in the spin relaxation rate and specific heat of the compound $CeAl_2$, and the frequency dependence of the relaxation time in $NaNO_2$ are discussed within this theory. There are several evidences that some of low temperature and low frequency properties, observed in quantum systems ordered in incommensurate phase, may be explained by the existence of new low frequency features in the spin (and pseudospins) excitation spectrum of the incommensurate structures at low temperatures. This conclusion represents the main new result.

1. Introduction

Excitation spectra and response functions expected to occur in quantum incommensurable modulated systems were studied in [1]-[7]. It was shown that the imaginary part of the transversal magnetic dynamic susceptibility decreases to zero linearly with frequency even in the case when a strong single ion easy anisotropy axis exists. This result is consequence of new low energy spin excitations which should be present in studied systems as it was predicted by theoretical nonperturbative approach to solve an infinity set of related coupled equations of motion. Theoretical energy spectrum of excitations in these systems is point-like due to incommensurability. Temperature fluctuations and effects of various defects are expected to wash out the smallest energy gaps in spectrum in real systems. Thus an energy spectrum represented by an envelope of the theoretical one is a reasonable approximation which should be compared with experimental one. The envelope curves preserve main characteristics of the point-like spectrum, namely the largest gaps, singularities with the largest weight, and the low energy excitations. The last mentioned property is a new feature which should be reflected in properties of corresponding materials at low temperatures.

Recently we applied the above mentioned concepts to several magnetic and dielectric systems. Let us present our main results for two of them:

[1]Presented at MECO (Middle European CoOperation) 19, Smolenice, Slovakia, April 11-15, 1994

1. $CeAl_2$: it is one of few magnetic quantum systems in which frozen-in incommensurable modulated transversal spin-wave was observed to exist as the ground state, [8]. Our related papers are concerning this material are: [9] and [10].
2. $NaNO_2$: Sodium nitrite crystallizes in the body-centered orthorhombic phase. It orders via sequence of order-disorder type of phase transitions, conveniently described by the appropriate quantum Ising pseudospin model. We concentrate on the temperature region between $T_i = 164^oC$ and $T_f = 162.5^oC$, where there exists a stable incommensurate modulated phase, [11]. The origin of this phase is due to competition of the antiferroelectric coupling between the next nearest neighbours and the ferroelectric coupling between the nearest neighbours. Our related paper concerning this material is: [12].

2. The dynamic equations

Our model description was based on the simplest exchange Hamiltonian leading to relevant description of modulated states. It has the form which is generalized from that introduced by Elliot, [13]:

$$H = -\int_{BZ} [\frac{1}{2} J(\mathbf{q})(m^+_{\mathbf{q}} m^-_{\mathbf{q}} + m^-_{\mathbf{q}} m^+_{\mathbf{q}}) + K(\mathbf{q}) m^x_{\mathbf{q}} m^x_{\mathbf{q}}] d^3q, \tag{1}$$

where $K(\mathbf{q}) \equiv J(\mathbf{q}) + D$. The interaction constant D is positive. The $J(\mathbf{q})$ interaction energy is such that together with strong local anisotropy D leads to an amplitude modulated state. The spins, $\mathbf{m}$, order with a modulation wavevector.

Let us now consider the Hamiltonian (1) which determines transition to the ordered modulated ground state of the spins $\mathbf{m}$. The integral in (1) is over the first Brillouin zone (BZ) of the paramagnetic phase and we assume that

$$\begin{aligned} J(\mathbf{q}) &\equiv J_1 \cos(\mathbf{qa}) + J_2 \cos(2\mathbf{qa}) + J_0(\mathbf{qb}, \mathbf{qc}), \\ m^\alpha_{\mathbf{q}} &\equiv \sum_{\mathbf{r}} \exp(i\mathbf{qr}) m^\alpha_{\mathbf{r}}. \end{aligned} \tag{2}$$

Here $\alpha = x$ and $\pm$, the lattice site position index is $\mathbf{r}$. The modulation direction vector $\mathbf{a}$ forms an orthogonal base together with remaining vectors $\mathbf{b}$ and $\mathbf{c}$. A strong effective uniaxial anisotropy energy $D \gg | J(\mathbf{q}) |$ favors an easy spin axis $\mathbf{x}$. In our case of $CeAl_2$ we have $\mathbf{a} \parallel (-110)$. Thus the transversal wave is formed at low temperatures. A random-phase-approximation type analysis of (1), analogous to that made in the longitudinal wave case in [8], tells us that if J($\mathbf{q}$) has a maximum at a wave vector $\mathbf{Q}$ with the amplitude $Q \equiv \mathbf{Qa}$ given by $\cos(Q) = -J_1/4J_2$ under appropriate values of the nearest neighbour and next nearest neighbour interaction constants J_1 and J_2, then there exists an amplitude modulated ground state

$$\begin{aligned} < m^x_{\mathbf{q}} > &= \frac{m}{2}(\exp(-i\phi).\delta(\mathbf{q} - \mathbf{Q}) + \exp(+i\phi).\delta(\mathbf{q} + \mathbf{Q})), \\ < m^x_{\mathbf{r}} > &= m \cos(\mathbf{Qr} + \phi), \\ < m^z_{\mathbf{r}} > &= m^y_{\mathbf{r}} = 0. \end{aligned} \tag{3}$$

Parameters J_1, J_2 and D are such that the mean field type stability condition

$$0 < \beta < 1/3 \tag{4}$$

is satisfied. Here

$$\begin{aligned} \beta &\equiv -J_2/D^*, \\ D^* &\equiv m(D + J_1 \cos(Q) + J_2 \cos(2Q)). \end{aligned} \tag{5}$$

ϕ is an arbitrary angle, m is the amplitude of the spin $\mathbf{m}$. We assume that the transversal interaction energies, which stabilize the three dimensional ordering, are small and we put $J_0 = 0$ in our following mean-field type approach.

The energy of the ground state is independent of the phase ϕ for any incommensurate phase. The later phase is characterized by $Q \neq 2\pi M/N$, where M and N are any integers. The wave (3) contains the basic harmonics only. Above the transition temperature $T_N = 3.8K$ the paraphase realizes. Far below the transition temperature firstly the squaring of the modulation of the wave was expected to become important and then a phase transition to a homogeneous phase, see in [13]. In this paper we restrict ourselves to consider only the single plane wave regime which was the only experimentally observed regime up till now in $CeAl_2$. As it was noted in [14] the weakness of the higher harmonics (and thus absence of the squaring) may be explained by the fairly constant moment value in the spiral structure.

As concerning the spin excitations above the ground state (3) it is known for a long time, [1]-[7], that the operator $m^+_{\mathbf{q}}$ or any finite combination of these operators with different wavevectors is not a normal coordinate in the incommensurate state of the system. To describe spin excitations in such situation we adopt here the procedure using continued fractions and developed in [4] - [7] for the longitudinal wave case. Let us consider wavevectors $\mathbf{q}$ and $\mathbf{Q}$ taken to be parallel. Denoting by

$$\begin{aligned} W_n(q) &\equiv D^*(1 + \alpha \cos(q + nQ) + \beta \cos(2q + 2nQ)), \\ \alpha/\beta &\equiv -4 \cos(Q), \end{aligned} \tag{6}$$

and the Fourier components of the spin operators by $m^+_{q+nQ} \equiv m^+_n$, where n is any integer, we have found an infinite set of coupled equations of motion

$$i\hbar\partial_t m^+_n = W_{n-1}(q) m^+_{n-1} + W_{n+1}(q) m^+_{n+1} \tag{7}$$

for the operators m^+_n. Theses equations are formally identical with the equations (7.5) in [7].

We have studied numerically the imaginary part of the magnetic susceptibility, Im $\chi^{+-}(q, \omega)$, for those values of the parameters α, β, M and N, for which an approximate description of the compound $CeAl_2$ below the Neél temperature is obtained

$$M = 2, N = 9, \alpha = -0.0893.$$

The imaginary part of the transversal magnetic susceptibility with parameters chosen to correspond to $CeAl_2$ is more thoroughly described and discussed in [10]. We have found that qualitatively all characteristics of the excitation spectrum are the same as those found in [4] - [7]. Quantitatively these characteristics correspond to our set of parameters. We emphasize the existence of bands of excitations, the existence of band edge singularities and the existence of new low frequency modes. It is easy to find that

$$\lim_{\omega\to 0}\frac{Im\chi^{+-}(q,\omega)}{\omega}=+\frac{m}{W_0^2(q)}, \tag{8}$$

where the spin amplitude m has its usual mean field square root type behaviour.

3. The spin-lattice relaxation rate (T_1^{-1})

Al nuclear-quadrupole-resonance studies, [9], of $CeAl_2$ at low temperatures reveal a linear temperature dependence of the spin-lattice relaxation rate (T_1^{-1}). This behaviour cannot be explained within existing theories. We argued that magnetic low-energy excitations of incommensurable modulated structure in $CeAl_2$ may account for a large fraction of linear behaviour of both quantities.

The NQR and NMR spectra are sensitive as well to the static magnetic structure (lineshape) as to the dynamic properties (spin relaxation time). This later quantity, $\frac{1}{T_1}$, may be calculated using the definition given in [15] for Al nucleus sites

$$\frac{1}{T_1}=\frac{k_BT}{\hbar N_{latt}}\sum_{q\nu}|A_\nu(q)|^2\frac{Im\chi^{\nu\nu}(q,\omega)}{\omega}, \tag{9}$$

where the resonance frequency $\omega\to 0$. Here A_ν is the Fourier transform of the hyperfine coupling, the sum on ν in (8) is over components of the diagonal ($\nu=x,y$), N_{latt} is the number of the lattice sites. We have found an explicit expression for (9) in the limit of low temperatures. It is sufficient in this limit to calculate the susceptibility without taking into account thermally activated processes due to temperature T prefactor in (9). The spin relaxation rate is thus found to be

$$\begin{aligned}T_1^{-1}&\approx T/\kappa,\\ \kappa^{-1}&\equiv k_B\hbar m\eta\omega_{hf}^2/D^*,,\\ \eta&\equiv\frac{1}{N_{latt}}\sum_q\frac{Im\chi^{+-}(q,\omega)}{m\omega}\Bigg|_{\text{dimensionless}},\end{aligned} \tag{10}$$

where $(\hbar\omega_{hf})^2\equiv<|A_\nu(q)|^2>$. For our set of parameters we have found $\eta=0.518$. Note, that the spin relaxation rate (10) depends linearly on temperature in those incommensurable systems in which the amplitude m and the modulation wavevector **Q** are only weakly temperature dependent,.This situation occurs also in the case of $CeAl_2$ compound below the critical temperature T_N but outside the critical region. The linear temperature behaviour in (10) should be compared to the exponential type behaviour found within the same model for the commensurate case [16].

The specific heat excess was calculated recently by the authors, [10], and was found to be linearly temperature dependent below the transition temperature. This corresponds with our experimental observation. [9].

4. Dielectric response

Response of modulated phases in materials with the order-disorder type of phase transition was studied recently. Description of dynamics by Bloch equations enabled us to calculate complex dynamic dielectric susceptibility. The complex Debye-relaxation behavior due to phase and amplitude excitations is found using perturbation approach up to the fourth order in the pseudospin amplitude, and confirmed by the nonperturbative envelope approach. Absorption peak shifts its maximum to lower frequencies when temperature is lowered, also its shape deforms and its height is lower than that when coupling of the homogeneous mode to higher order modes is turned off. Low frequency behavior of the real part of the susceptibility is modified.

It is well known that the effective relaxation inverse time for any system may be expressed using real and imaginary part of the susceptibility:

$$\frac{1}{\tau(\omega)}=\frac{\omega\chi'}{\chi''}.$$

Temperature and frequency dependence of the effective inverse relaxation time for $NaNO_2$ was recently calculated using perturbative analytical approach and using envelope methods, [12]. We have found temperature and frequency dependencies of the effective inverse relaxation time for $NaNO_2$ which are not consistent with the single relaxation process scenario. Both quantities are frequency independent in the simple Debye relaxators. The incommensurate structures display complex frequency dependent behavior of both quantities due to coupling between modes. Such a complex behaviour was observed in several dielectrics with incommensurate phases. Our results point to existence of a transition region in frequency dependece of the effective relaxation times between two different regimes (high and low frequency regions). Moreover temperature dependence of the inverse relaxation time shows a small peak immediately below the transition temperature followed by a steep decrease. This curve shifts up for higher frequencies. Hatta [11], observed qualitatively the same frequency and temperature dependence of the relaxation times in $NaNO_2$. This correspondence is may be improved taking more complicated but more realistic model into considerations.

5. Discussion

Both linear dependencies, in the specific heat and the spin-lattice relaxation time, were recently observed in [9] below T_N outside the critical temperature region. Thus we conclude that at least a part of the observed linear dependence in the spin-lattice relaxation rate and in the specific heat of the compound $CeAl_2$ may be explained by existence of new low frequency modes in the spin excitation spectrum of the incommensurate magnetic structure. While this structure seems to be non-chiral spiral, we would like to note that the predicted low frequency and low temperature behaviour should be expected to be present also in other magnetic structures (which may be realized even in $CeAl_2$) whenever they are incommensurable modulated. The results of this paper contribute to better understanding of such structures. Experimental confirmation of presence or of absence of their new thermodynamic and dynamic properties, such as predicted in our paper, may serve as a strong test of the theory.

Temperature and frequency dependence of the effective inverse relaxation time and the effective Curie constant for $NaNO_2$ were calculated in our paper recently. Both quantities are frequency independent in the simple Debye relaxators. The incommensurate structures display complex frequency dependent behavior of both quantities due to coupling between modes. Such a complex behaviour was observed in several dielectrics with incommensurate phases, f.e. Hatta observed frequency dependence of the relaxation times in $NaNO_2$.

Thus we can summarize results from above in this contribution, to find a new result: there are several evidences that some of low temperature and low frequency properties, observed in quantum systems ordered in incommensurate phase, may be explained by the existence of new low frequency features in the spin (and pseudospins) excitation spectrum of the incommensurate structures at low temperatures. As evidence here we mentioned the observed linear dependence in the spin relaxation rate and specific heat of the compound $CeAl_2$, and the frequency and temperature dependence of the relaxation time in $NaNO_2$.

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