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Spin anharmonicity and zero-point fluctuations in weak itinerant electron magnets

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Abstract. It is shown that the conventional spin fluctuation (SF) theory of weak itinerant magnets does not take into account the effects of strong spin anharmonicity caused by large zero-point SF amplitudes and, therefore, may break down in anharmonic magnets. A microscopic approach based on a quasiparticle Fermi liquid description of strongly anharmonic weak itinerant magnets is presented generalizing the conventional theory to account for both thermal and zero-point SF.

Weak itinerant electron magnets close to a magnetic instability are usually described within the spin fluctuation (SF) theory which is believed to be well established on the microscopic basis related to the Hubbard model [1, 2] or Fermi liquid concept [3]. However, the main approximations of the SF theory remain unclear. In the present paper we show that the most crucial point of the conventional SF theory is due to the neglect of the zero-point SF effects, which corresponds to the weak-spin-anharmonicity limit. Recently we presented a theory accounting for the zero-point effects in strongly anharmonic itinerant magnets, which was based on the phenomenological quantum Ginzburg–Landau model [4]. Here we formulate a microscopic Fermi liquid approach accounting for the strong spin anharmonicity effects caused by large zero-point SF amplitudes in weak itinerant magnets.

We start with the following model for the matrix elements of the Fourier transform of a quasiparticle energy $\hat{\epsilon}(\omega)$ for a ferromagnetic Fermi liquid [5]:

$$\langle \sigma, \mathbf{p} + \hbar \mathbf{k} | \hat{\epsilon} | \sigma', \mathbf{p} \rangle = 2\pi \delta_{\sigma\sigma'} \delta_{\mathbf{k}0} \delta(\omega) \epsilon^\sigma(\mathbf{p}) + \hat{\sigma}_{\sigma\sigma'} \Psi(\mathbf{k}) m(\omega, \mathbf{k}) + \delta_{\sigma\sigma'} \phi_{\text{eff}}(\omega, \mathbf{k}) n(\omega, \mathbf{k}) \quad (1)$$

which are assumed to be linear in the amplitudes $m(\omega, \mathbf{k})$ and $n(\omega, \mathbf{k})$ of spin and density fluctuations with a frequency ω and wavevector \mathbf{k} . Here $\epsilon^\sigma(\mathbf{p})$ is the equilibrium energy of quasiparticles without taking into account fluctuation effects, $\sigma = \pm 1$, and \mathbf{p} denote the spin and momentum, $\hat{\sigma}$ are the Pauli matrices and $\Psi(\mathbf{k})$ describes the exchange interaction of quasiparticles. The function $\phi_{\text{eff}}(\omega, \mathbf{k})$ accounts for the Coulomb interaction of quasiparticles and its dynamical screening by the crystal lattice. Below we set $\phi_{\text{eff}}(\omega, \mathbf{k}) \rightarrow \infty$ neglecting the coupling of SF to the lattice and electron density fluctuations (see [5]). Within this model the free energy treated as a function of the magnetization M and temperature T may be written in the conventional form [1–3]

$$F(M, T) = F_0(M, T) + \Delta F(M, T). \quad (2)$$

Here $F_0(M, T)$ is the Hartree–Fock, or Stoner free energy, which for weak magnets may be expanded in terms of M ,

$$F_0(M, T) = F_0(T) + \frac{1}{2\chi_0} M^2 + \frac{\gamma_0}{4} M^4 + \dots \quad (3)$$

with the coefficients $\chi_0 = \nu/(1 + \Psi\nu)$ and $\gamma_0 = (3\nu'^2 - \nu\nu'')/6\nu^5$, where ν, ν', ν'' are the quasiparticle density of states and its derivatives at the Fermi level, and $\Psi = \Psi(0)$ (we use the system of units with $\mu_B = 1$, where μ_B is the Bohr magneton). The contribution $\Delta F(M, T)$ accounts for the SF effects and within the Fermi liquid model (1) may be written in the form (cf [1] and [6])

$$\Delta F = 2\hbar \sum_{\nu=t,l} \sum_{\omega, \mathbf{k}} \left(N_\omega + \frac{1}{2} \right) \int_0^{\Psi(\mathbf{k})} d\Psi \operatorname{Im}[\chi_\nu(\omega, \mathbf{k}) - \chi_\nu^{(0)}(\omega, \mathbf{k})] \quad (4)$$

where

$$\sum_{\omega, \mathbf{k}} = \int_0^{+\infty} \frac{d\omega}{2\pi} \sum_{\mathbf{k}}.$$

Equation (4) can be split into a thermal part containing $N_\omega = [\exp(\hbar\omega/k_B T) - 1]^{-1}$ and a contribution due to zero-point SF. Here $\chi_\nu^{(0)}(\omega, \mathbf{k})$ are the transverse ($\nu = t$) and longitudinal ($\nu = l$) dynamical magnetic susceptibilities of a Fermi liquid at $\Psi = 0$ and $\phi_{\text{eff}}(\omega, \mathbf{k}) = \infty$ (see, e.g., [5]). We treat the finite Ψ dynamical susceptibilities $\chi_\nu(\omega, \mathbf{k})$ in the spirit of the Moriya–Kawabata theory [1] as functions of M and take them in the conventional form

$$\chi_\nu^{-1}(\omega, \mathbf{k}) = \chi_\nu^{(0)-1}(\omega, \mathbf{k}) + \Psi + \lambda_\nu(\omega, \mathbf{k}). \quad (5)$$

Here the functions $\lambda_\nu(\omega, \mathbf{k})$ describe the effects of SF and vanish when these are neglected. When $\lambda_\nu(\omega, \mathbf{k}) \rightarrow 0$ equation (5) gives the magnetic susceptibilities [5] for the Fermi liquid model (1) with $\phi_{\text{eff}}(\omega, \mathbf{k}) = \infty$ and the SF effects neglected [5]. This is also related to the well known random phase approximation (RPA) [1, 6]. In the long-wavelength low-frequency limit we may write

$$\lambda_\nu(\omega, \mathbf{k}) \simeq \lambda_\nu(0, 0) = \lambda_\nu \quad (6)$$

where λ_ν are defined self-consistently by the equations

$$\lambda_t = \frac{1}{M} \frac{\partial \Delta F}{\partial M} \quad (7)$$

$$\lambda_l = \frac{\partial^2 \Delta F}{\partial M^2} \quad (8)$$

following from the coincidence of the static homogeneous susceptibilities $\chi_\nu(0, 0) \equiv \chi_\nu$ and the thermodynamic ones.

The approximation of the conventional SF approach, besides (6), is to neglect Ψ and M dependences of λ_ν and, following the Moriya–Kawabata theory [1], to set

$$\left| \frac{\partial \lambda_\nu}{\partial \Psi} \right| \ll 1 \quad (9)$$

$$\left| \frac{\partial \lambda_\nu}{\partial M} \right| \ll \frac{\partial (\chi_\nu^{-1})}{\partial M}. \quad (10)$$

Below we show that both these inequalities are related to the weak-spin-anharmonicity limit and do not hold in real weak itinerant magnets where large zero-point SF amplitudes give rise to strong spin anharmonicity effects.

Taking account of (9) the integration over Ψ in (4) yields the familiar result for the SF free energy [1–3], which in the rotationally invariant form is given by

$$\begin{aligned} \Delta F &= 2\hbar \sum_{\nu} \sum_{\omega, \mathbf{k}} \left(N_{\omega} + \frac{1}{2} \right) \text{Im} \{ \ln [1 + (\Psi(\mathbf{k}) + \lambda_{\nu}) \chi_{\nu}^{(0)}(\omega, \mathbf{k})] - \Psi(\mathbf{k}) \chi_{\nu}^{(0)}(\omega, \mathbf{k}) \} \\ &\equiv \sum_{\nu=1} \Delta F_{\text{RPA}}^{(\nu)}(\lambda_{\nu}) \end{aligned} \quad (11)$$

and in the limit $\lambda_{\nu} = 0$ is related to the RPA free energy of the paramagnon theory [6]. Taking account of the inequality (10) equations (7), (8) and (11) yield the following solution, e.g., for λ_t (cf [1–3]):

$$\lambda_t = \lambda_t = \gamma_0 \{ 2[(\delta m_t^2)_{\text{zp}} + (\delta m_t^2)_{\text{T}}] + 3[(\delta m_t^2)_{\text{zp}} + (\delta m_t^2)_{\text{T}}] \} \quad (12)$$

where

$$(\delta m_{\nu}^2)_{\text{zp}} = 2\hbar \sum_{\omega, \mathbf{k}} \text{Im} \chi_{\nu}(\omega, \mathbf{k}) \quad (13)$$

and

$$(\delta m_{\nu}^2)_{\text{T}} = 4\hbar \sum_{\omega, \mathbf{k}} N_{\omega} \text{Im} \chi_{\nu}(\omega, \mathbf{k}) \quad (14)$$

are the zero-point and thermal contributions to the averaged squared amplitudes of transverse and longitudinal SF. In the conventional SF treatment [1–3] zero-point SF contributions to (12) are assumed to be temperature and magnetization independent, which allows us to incorporate them into the exchange parameter $\Psi(\mathbf{k})$.

However, the recent neutron scattering experiments in weak itinerant magnets [7–9] presented direct evidence for large zero-point SF amplitudes strongly dependent on temperature. This was also anticipated by Takahashi [10] who accounted for the effects of the zero-point SF amplitude variation basing his calculation on the constraint of the approximate conservation of the total local magnetic moment. However, this constraint is not borne out by thermodynamics for weak itinerant magnets [4].

To illustrate these effects we may use the conventional form for the dynamical susceptibilities [1, 2], $\chi_{\nu}^{-1}(\omega, \mathbf{k}) = \chi_{\nu}^{-1} + c\mathbf{k}^2 - i\omega/\Gamma k$, provided $\omega \leq \omega_c$ and $k \leq k_c$ are in the paramagnon region. Here c and Γ describe the spatial and time dispersions, $\omega_c \simeq kv_F$ and k_c are the cut-off frequency and wavevector, v_F is the Fermi velocity. As was shown in neutron scattering experiments [9] this form may hold over the whole Brillouin zone up to the frequencies $\omega \sim 8k_B T_C/\hbar$, where T_C is the Curie temperature. We assume that the inverse susceptibilities are small enough,

$$\chi_{t,1}^{-1} \ll ck_c^2 \quad (15)$$

not only near T_C but also at low temperatures (see the discussion below) and expand the squared zero-point SF amplitudes (13) in powers of χ_{ν}^{-1} (cf [10]),

$$(\delta m_{\nu}^2)_{\text{zp}} = \delta m_c^2 - (g_0/\gamma_0)\chi_{\nu}^{-1} + \dots \quad (16)$$

Here $\delta m_c^2 \sim \hbar \Gamma k_c^4$ is the squared amplitude of the zero-point SF at $\chi_v^{-1} = 0$ and

$$g_0 = 4\hbar\gamma_0 \sum_{\omega, \mathbf{k}} [\operatorname{Re} \chi_v(\omega, \mathbf{k}) \operatorname{Im} \chi_v(\omega, \mathbf{k})]_{\chi_v^{-1}=0} \sim \frac{\gamma_0 \delta m_c^2}{ck_c^2} \quad (17)$$

is the spin anharmonicity parameter [4, 11] related to the zero-point SF. We want to emphasize that the derivation of equations (16) and (17) does not depend on the precise form for the dynamical susceptibilities used above and has a wide range of validity. Using the estimates

$$\left| \frac{\partial \lambda_v}{\partial \Psi} \right| \sim \left| \frac{\partial \lambda_v}{\partial M} \right| \left/ \frac{(\partial \chi_v^{-1})}{\partial M} \right. \sim g_0 \quad (18)$$

following from (12) and (16) we conclude that inequalities (9) and (10) hold in the weak-SF-coupling limit, when the spin anharmonicity parameter (17) is small, $g_0 \ll 1$, which actually is the main approximation of the conventional SF theory. As we have recently shown [4, 11], this approximation does not hold for the weak itinerant magnets MnSi, Ni₃Al and ZrZn₂. We also suspect that this is a general situation for itinerant magnets unlike, e.g., anharmonic crystals [12] where the anharmonicity parameter is usually small.

Here we have to generalize the conventional SF theory [1–3] to account for the large spin anharmonicity effects caused by the zero-point SF. As was shown above, the role of these effects is twofold. First, they give rise to the variation of zero-point SF amplitudes, which may essentially affect thermal properties of itinerant magnets. Second, they may lead to the violation of the inequalities (9) and (10) and to the breakdown of the RPA expression (11) for the SF contribution to the free energy. Below we take into account both of these aspects of spin anharmonicity and calculate the free energy using a variational procedure instead of a perturbation one without any restrictions on the spin anharmonicity parameter g_0 .

In our description we must allow for the M and Ψ dependences of $\lambda_v = \lambda_v(M, \Psi)$ and account for the finite derivatives $\partial \lambda_v / \partial M \neq 0$, $\partial \lambda_v / \partial \Psi = \xi_v \neq 0$ which are usually neglected [1–3]. These derivatives are strongly affected by spin anharmonicity due to the zero-point SF and according to the estimate (18) vanish in the weak-coupling limit, $g_0 \rightarrow 0$. Here we assume that ξ_v are functions of the spin anharmonicity parameter g_0 only and neglect their Ψ and M dependences, which yields the following model for λ_v :

$$\lambda_v = \lambda_{v0}(M) + \xi_v \Psi. \quad (19)$$

This assumption agrees with the estimates (18) and below it will be verified via the variational procedure.

From (4), (5) and (19) follows the explicit formula

$$\Delta F = \sum_v \frac{1}{1 + \xi_v} \Delta F_{\text{RPA}}^{(v)}(\lambda_v) \quad (20)$$

for the SF contribution to the free energy, where $\Delta F_{\text{RPA}}^{(v)}$ is given by (11). We emphasize that this formula accounts for the M and Ψ dependences of λ_v and differs from the conventional RPA like result (11) by the factors $(1 + \xi_v)^{-1}$ describing anharmonic effects beyond the weak-coupling approximation of the existing theory [1–3].

In principle, equations (5)–(8) and (20) form a complete set of equations to calculate the functions $\lambda_\nu(M, \Psi)$ and $\xi_\nu(g_0)$. However, this task is rather complicated and we simplify it by expanding (20) in powers of the inverse susceptibilities χ_ν^{-1} . Similarly to (16) we have

$$\Delta F = \frac{1}{2} \sum_{\nu=i,1} \frac{1}{1 + \xi_\nu} \left[(\delta m_c^2 + \delta m_T^2) \chi_\nu^{-1} - \frac{g_0}{2\gamma_0} \chi_\nu^{-2} + \dots \right]. \quad (21)$$

Here $\delta m_T^2 = k_B T k_T / 2\pi^2 c$ is the thermal contribution (14) to the squared SF amplitude at $\chi_\nu^{-1} = 0$, and $k_T = (T/T_m)^{1/3} k_c$ is the characteristic wavevector of thermal SF in the quantum temperature range $T \ll T_m$, where $k_B T_m \sim \hbar \Gamma c k_c^3$ is the maximum energy of SF which in typical weak itinerant magnets is much greater than $k_B T_C$ [2]. The terms containing the spin anharmonicity parameter g_0 account for the variation of the zero-point SF amplitudes (13). In equation (21) we have neglected the constant contribution independent of $\chi_{i,1}$ and the Ginzburg–Landau term $\sim k_B T (c \chi_\nu)^{-3/2}$ which is not important outside the critical region [4, 11] and is small when

$$\chi_\nu^{-1} \ll c k_T^2. \quad (22)$$

Inequalities (15) and (22) define the range of validity of the expansion (21) and do not necessarily require the anharmonicity parameter g_0 to be small (see the discussion below).

On the other hand, the SF free energy (21) may be also expanded in terms of the magnetization M provided the Ginzburg–Levanyuk criterion holds and the Landau theory of phase transitions is applicable. This allows us to find the solution of equations (5)–(8) and (21) expanding $\lambda_\nu(M)$ in powers of M . Retaining terms up to the second order in M we have

$$\lambda_\nu(M) = -5g\nu^{-1} + 5\gamma(\delta m_c^2 + \delta m_T^2) + (\gamma - \gamma_0)M^2 \quad (23)$$

$$\lambda_\nu(M) = -5g\nu^{-1} + 5\gamma(\delta m_c^2 + \delta m_T^2) + 3(\gamma - \gamma_0)M^2 \quad (24)$$

where g and γ are the renormalized spin anharmonicity parameter and SF coupling constant given, respectively, by

$$g_0 = g \frac{1 + 6g}{1 - 5g} \quad (25)$$

and

$$\gamma = \gamma_0 \frac{1 - 5g}{1 + 6g}. \quad (26)$$

We also find the parameters

$$\xi_i = \xi_1 = -5g = \xi \quad (27)$$

which are independent of Ψ , verifying the model (19) used above. Substituting relations (23)–(26) into equations (5) and (21) we obtain the free energy (2) in the Landau form,

$$F(M, T) = F_0 + \frac{3}{2(1 + \xi)\chi(T)} \left[\delta m_c^2 + \delta m_T^2 - \frac{g}{2\gamma\chi(T)} \right] + \frac{1}{2\chi(T)} M^2 + \frac{\gamma}{4} M^4 \quad (28)$$

where

$$\chi^{-1}(T) = \chi_0^{-1}(1 - 5g) + 5\gamma(\delta m_c^2 + \delta m_T^2). \quad (29)$$

A similar result may be obtained within the quantum Ginzburg–Landau model [4]. It follows from (25)–(28) that the anharmonic effects arising from the variation of the zero-point SF amplitudes (16) and from the Ψ dependence of λ_ν given by (19) are equally important and are governed by the same parameter $g = g(g_0)$.

Substituting the free energy (28) into the conventional equations of state, $B = (\partial F/\partial M)_V$ and $P = -[\partial(FV)/\partial V]_{MV}$, which define the magnetization M and volume V as functions of the magnetic field B and pressure P , we arrive at the magnetic equation of state

$$\frac{B}{M} = \chi^{-1}(0) + \gamma(M^2 + 5\delta m_T^2) \quad (30)$$

and the magnetic contribution of the volume strain

$$\omega_m = \frac{C_0}{K}(M_L^2)_{\text{tot}} = \frac{C}{K}(M_L^2)_T + 3\frac{C_0}{K}[\delta m_c^2 - g/\chi(0)\gamma]. \quad (31)$$

Here K is the bulk modulus, $C_0 = -\frac{1}{2}V^2\partial(\chi_0 V)^{-1}/\partial V$ and $C = -\frac{1}{2}V^2\partial(\chi(0)V)^{-1}/\partial V \simeq C_0(1 - 5g)$ are the magnetoelastic coupling constants, unrenormalized and renormalized by zero-point SF, respectively and

$$(M_L^2)_{\text{tot}} = M^2 + \sum_\nu [(\delta m_\nu^2)_{\text{zp}} + (\delta m_\nu^2)_T] \quad (32)$$

and

$$(M_L^2)_T = M^2 + 3\delta m_T^2 \quad (33)$$

are the local squared magnetic moments with and without the account of zero-point SF. In the above formulae we neglected the volume dependences of the SF coupling constant γ and of the spatial and time dispersions of the dynamical spin susceptibilities.

We now discuss our results with respect to the predictions of the conventional SF theory [1–3]. First, we emphasize that the equations of state (30) and (31) have essentially the same form as was found previously without zero-point SF, provided the temperature independent contribution to ω_m caused by zero-point SF is neglected. Therefore, it is not possible to distinguish our results from the conventional ones, discussing only the magnetic or magnetovolume measurements.

However, the parameters $\chi^{-1}(0)$, γ and C in (30) and (31) incorporate the effects of zero-point SF and differ from the initial ones, χ_0^{-1} , γ_0 and C_0 which enter the results of the conventional theory. In the weak-SF-coupling limit, when $g \simeq g_0 \ll 1$, $\gamma \simeq \gamma_0$ and $C \simeq C_0$ equations (29)–(31) yield the magnetic equation of state [2, 3] and expression for the magnetovolume effect [1] arising in the conventional SF theory, except for the constant contributions to $\chi^{-1}(0)$ and ω_m , containing the squared zero-point SF amplitudes δm_c^2 . According to (25) and (26) the effects of spin anharmonicity due to zero-point SF reduce the SF and magnetoelastic coupling constants which vanish, $\gamma \sim \gamma_0/g_0$ and $C \sim C_0/g_0$, in the limit of strong spin anharmonicity, when $g_0 \gg 1$ and $g \simeq \frac{1}{5}$. The Stoner criterion, $\chi^{-1}(0) < 0$, with $\chi^{-1}(0)$ given by (29) is also modified with respect to the zero-point

SF effects which tend to suppress a ferromagnetic instability by reducing the negative contribution $\sim \chi_0^{-1}$ and adding the positive term $\sim \delta m_c^2$.

We also point out that the formulae presented above establish the link between the parameters χ^{-1} , γ and C which define the magnetic and magnetovolume properties of weak itinerant magnets and may be directly inferred from experiments, and the constants χ_0^{-1} , γ_0 and C_0 related to the band structure calculations (see, e.g., [13]). Our analysis which is based on a microscopic approach also gives some insight into the recent phenomenological description of thermal properties of weak itinerant magnets [1, 2]. Although the authors of [1, 2] formally neglected the zero-point SF effects they used the empirical parameters $\chi^{-1}(0)$ and γ which as we show actually incorporate zero-point SF effects. This explains the success of the quantitative interpretation of the finite-temperature properties of a series of metals [1, 2], which is not obvious in view of the temperature dependent effects induced by zero-point SF.

Finally, we discuss the validity of our approach which besides the well known Ginzburg–Levanyuk criterion is limited by the inequalities (15) and (22) allowing us to expand the SF contribution to the free energy (21) in terms of χ_v^{-1} . Using the magnetic equation of state (30) and the estimate for δm_T^2 presented above we find from (15) that the present description is valid even at $T = 0$ provided the coefficient $\chi^{-1}(0)$ and the spontaneous magnetization M_0 are small enough, and the Curie temperature T_C is sufficiently low

$$\frac{|\chi^{-1}(0)|}{ck_c^2} \sim 5g \frac{M_0^2}{\delta m_c^2} \sim 5g \left(\frac{T_C}{T_m} \right)^{4/3} \ll 1. \quad (34)$$

Similarly it follows from (22) that our approach is valid not only near T_C , when $|T - T_C| \ll T_C$, but also far above T_C provided

$$5g \left(\frac{T}{T_m} \right)^{4/3} \ll 1. \quad (35)$$

We emphasize that inequalities (34) and (35) not only are satisfied in the weak-anharmonicity limit, $g_0 \ll 1$, but also hold in strongly anharmonic itinerant magnets where $g_0 \gg 1$ and $5g \simeq 1$, provided $T_m \gg T_C$.

The zero-point SF effects discussed here play an important role in weak itinerant magnets as was shown by the recent neutron scattering investigations [7–9] where large zero-point SF amplitudes, $\delta m_c \sim 1$ (in μ_B per magnetic atom), were observed. Using the magnetic and neutron scattering data from [2] and [8] we can estimate the effects of anharmonicity caused by zero-point SF, e.g. for MnSi. Integrating (17) with the parameters [2, 8] $c = 0.21 \times 10^5 \text{ \AA}^2$, $k_c = 0.86 \text{ \AA}^{-1}$ and $\hbar\Gamma = 2.6 \text{ \mu eV \AA}$ which describe the spatial and time dispersions of the dynamical susceptibilities and using the measured constant [2] $\gamma = 0.15 \text{ G}^{-2}$ we estimate the renormalized spin anharmonicity parameter [11] $g = 0.18$. Then from (25) and (26) we calculate the unrenormalized quantities $g_0 = 5.4$ and $\gamma_0 = 4.9 \times 10^{-3} \text{ G}^{-2}$. These estimates show the importance of the zero-point SF effects which give rise to the strong spin anharmonicity not accounted for by the conventional SF theory of weak itinerant magnetism.

To conclude, we have presented a new microscopic approach to describe the zero-point SF effects in anharmonic weak itinerant magnets. We showed that the two aspects of spin anharmonicity, one related to the temperature variation of zero-point SF amplitudes and the other giving rise to the anharmonic effects beyond the weak-coupling approximation are of equal importance for weak itinerant electron magnets.

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