Quasielastic magnetic scattering of neutrons at the systems with heavy fermions

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(March 16, 2018)

The theory of the quasielastic magnetic scattering of neutrons at the spin liquid with RVB (resonance valence bonds) correlations is presented. Calculations demonstrate that the dependence of the scattering cross section on the energy transfer reproduces experimental shape of quasielastic peak in heavy fermion systems. It is shown that Fermi statistics of the spin liquid elementary excitations leads to the oscillations of the quasielastic scattering total cross section as a function of the momentum transfer.

Accepted to JETP Lett., Vol. 68

1. Since it was suggested [1,3] to treat the state with heavy fermions (HF) as the spin liquid (SL) with RVB type of spin-spin correlations the validity of RVB conception is still the subject of considerable discussions. Although conception of RVB type SL appeared to be fruitful for quantitative description of the thermodynamic properties and low energy spectral [5] properties of HF systems there is no unambiguous proof of existence of RVB correlations. The basic stumbling-block on the way to the SL identification is the lack of decisive experiment which can give possibility to accept or reject RVB conception.

According to the RVB scenario the system of localized spins transforms at temperatures close to Kondo temperature \( T_K \) into the half filled band of magnetic excitations with the bandwidth \( T^* \sim T_K \) [3,6,7]. The basic fingerprint of the RVB correlations, which distinguishes SL state from the system of localized spins, is the Fermi statistics of the elementary excitations. Therefore, the study of consequences of the different statistical distributions which characterize SL and localized spin regime is a proper way to reveal some properties which are peculiar only to the highly correlated RVB state.

In the present paper I suggest the theory of the quasielastic magnetic scattering of neutrons at the RVB type SL and show that change of the Boltzmann statistics of localized spins to the Fermi statistics of the SL excitations results in the oscillatory behavior of the total quasielastic cross section as a function of momentum transfer.

2. The basic model describing the HF state is the Anderson lattice Hamiltonian of the \( f \)-ions which are hybridized with the conduction electrons. The Coqblin - Schrieffer canonical transformation [3,4] eliminates the hybridization term and the lowest crystal field doublet \( \sigma = \pm \) can be treated in terms of exchange interaction between the spin states of different sites \( \mathbf{m} = 1, \xi (1 \) is the number of the elementary cell, \( \xi \) is the basis vector)\[H_{ex} = \sum_{\mathbf{m} \neq \mathbf{m}'} \sum_{\sigma \sigma'} f_{\mathbf{m} \sigma}^\dagger f_{\mathbf{m} \sigma'}, f_{\mathbf{m}' \sigma'}^\dagger f_{\mathbf{m}' \sigma} = 1 \]

Here \( f_{\mathbf{m} \sigma} \) (\( f_{\mathbf{m} \sigma}^\dagger \)) are creation (annihilation) Fermi operators of the spin state \( \sigma \) at the site \( \mathbf{m} \) which are subject to the constraint condition \( \sum_{\sigma} f_{\mathbf{m} \sigma} f_{\mathbf{m} \sigma}^\dagger = 1 \).

In the mean field approach the RVB state at low temperatures is characterized by nonzero intersite averages \( \langle f_{\mathbf{m} \sigma}^\dagger f_{\mathbf{m}' \sigma} \rangle \) and effective Hamiltonian of the SL low temperature states takes the form
\[H_{ex}^f = \sum_{\mathbf{m} \neq \mathbf{m}'} \sum_{\sigma \sigma'} A_{\mathbf{m} \mathbf{m}'} f_{\mathbf{m} \sigma}^\dagger f_{\mathbf{m}' \sigma}, \quad (2)\]

which formally resembles tight binding approximation for band spectrum. The constants \( A_{\mathbf{m} \mathbf{m}'} \) are determined by the values of exchange integrals \( I_{\mathbf{m} \mathbf{m}'} \) and anomalous averages [3]. The chemical potential \( \mu \) in the mean field approximation can be deduced from the global constraint for \( N \)-ion lattice \( N^{-1} \sum_{\mathbf{m}} \sum_{\sigma} \langle f_{\mathbf{m} \sigma}^\dagger f_{\mathbf{m} \sigma} \rangle = 1 \). Therefore, the RVB SL in the mean field approximation can be considered as a half filled band of Fermi quasiparticles with the width \( T^* \). The eigenstates \( | \lambda \rangle \) of the Hamiltonian (3) can be expressed in terms of eigenvectors \( \Xi_{\lambda \sigma}^\lambda \) in the form of superposition [3]

\[| \lambda \rangle = \sum_{\kappa \xi} \exp(i\kappa \lambda) \Xi_{\xi \sigma}^\lambda \varphi(r - 1 - \xi) | \sigma \rangle, \quad (3)\]

(Here \( \varphi(r - 1 - \xi) \) is the spatial wave function of localized \( f \)-electron; \( | \sigma \rangle \) is the spin component of the wave function and \( k_\lambda \) is the wave vector of the state \( | \lambda \rangle \).) The occupation numbers of the states \( | \lambda \rangle \) with the energies \( \varepsilon_\lambda \) at temperature \( T \) are described by Fermi distribution \( n_\lambda = (1 + \exp[(\varepsilon_\lambda - \mu)/T])^{-1} \).

3. The standard expression for the scattering function [10] per one magnetic ion with the momentum transfer \( \kappa \) and energy transfer \( \hbar \omega \) can be expressed in terms of the matrix elements of operator \( Q = \sum_{\kappa} \exp(i\kappa(1 + \xi)) \hat{J} \) and Cartesian components of unit vectors \( \hat{e}_\alpha = \kappa_\alpha/|\kappa| \)

\[S(\kappa, \hbar \omega) = b^2 N^{-1} \sum_{\alpha \beta} (\delta_{\alpha \beta - \kappa_\alpha \kappa_\beta}) \times \sum_{\lambda \lambda'} \Omega_{\lambda \lambda'} \langle \lambda | \hat{Q}_\alpha^\dagger \lambda' \langle \lambda' | \hat{Q}_\beta \lambda \rangle \delta(\hbar \omega - \varepsilon_{\lambda'} + \varepsilon_{\lambda}) \quad (4)\]

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(b is the magnetic scattering length \[10\]; \(\hat{J}\) is the operator of total magnetic moment). In case of the scattering at the localized crystal field states the statistic function \(\Omega_{\lambda^{'},\lambda^{'}}\) is connected with the Boltzmann distribution and depends only on the occupation numbers of the initial states \(\lambda\). On the other hand in case of Fermi statistics of SL excitations the occupation of the final state is also important and statistic function takes the form

\[
\Omega_{\lambda^{'},\lambda} = n_{\lambda}(1 - n_{\lambda^{'}}). \tag{5}
\]

Inserting the eigenstates \((3)\) into expression \((4)\) and neglecting the intesite matrix elements of operator \(\hat{J}\) between highly localized \(f\)-electron states one gets the following expression for the scattering function

\[
S_{sl}(\kappa, \hbar \omega) = B(\kappa) \sum_{\alpha \beta} (\delta_{\alpha \beta} - \hat{k}_{\alpha} \hat{k}_{\beta}) \times \sum_{\lambda \lambda^{'}} \Omega_{\lambda^{'},\lambda} \Theta_{\lambda^{'},\lambda}^{\alpha \beta}(\kappa) \delta(\hbar \omega - \varepsilon_{\lambda^{'}} + \varepsilon_{\lambda}), \tag{6}
\]

where

\[
\Theta_{\lambda^{'},\lambda}^{\alpha \beta}(\kappa) = I_{\lambda^{'},\lambda}^{\alpha}(\kappa) \left( I_{\lambda,\lambda^{'}}^{\beta}(\kappa) \right)^{*}, \tag{7}
\]

and

\[
I_{\lambda,\lambda^{'}}^{\alpha}(\kappa) = \sum_{\xi} \exp(-i\kappa \xi) \sum_{\sigma \sigma^{'}} (\Xi_{\xi,\sigma}^{\lambda})^{*} \Xi_{\xi,\sigma^{'}}^{\lambda^{'}}(\sigma | \hat{J}_{\alpha} | \sigma^{'}). \tag{8}
\]

Here \(B(\kappa) = b^{2} (g_{J} F(\kappa)/2)^{2}\) where \(g_{J}\) is the Lande factor and \(F(\kappa)\) is the formfactor of the magnetic ion.

4. First of all it have to be noted that dependence of the scattering function \((1)\) on the energy transfer \(\hbar \omega\) and temperature qualitatively describes the experimentally situation with the quasielastic peaks in HF systems \([1]\). The quasielastic peakshape in HF systems is treated usually by use of the phenomenological expression \((11)\)

\[
S_{sl}(\kappa, \hbar \omega = 0) = \frac{R(\kappa)}{2} \exp \left( \frac{\hbar \omega}{2T} \right) \sinh^{-1} \left( \frac{|\hbar \omega|}{2T} \right) \times \ln \left\{ \frac{1 + \cosh \left[ \frac{T^{*}/T}{1 + \cosh \left[ (T^{*} - |\hbar \omega|)/T \right]} \right]}{-1} \right\}. \tag{11}
\]

As may be seen from Fig. 1 the energy and temperature dependence of expression \((11)\) is in qualitative agreement with experimental measurements of quasielastic response in HF systems. Moreover, the energy dependence \((11)\) is similar for \(\hbar \omega < T^{*}\) to the phenomenological function \((1)\). It is instructive that generally accepted for the experimental data treatment fit of the formula \((10)\) to the scattering function \((11)\) gives \(\Gamma(T = 0) \sim T^{*}\). It have to be noted that breaks of the energy dependence at \(\hbar \omega = 0\) and restriction of \(S_{sl}(\kappa, \hbar \omega)\) by the energy range \([-2T^{*}, 2T^{*}\)] can be avoided by use of less singular density of states and involving the damping of SL excitations, respectively.

5. The energy dependence of the quasielastic scattering at the SL can not be used as a decisive argument in favor of RVB model applicability for HF systems because there are lot of models which give the same result (e.g. \([12, 13]\)). However, much more bright consequence of the RVB model, which can serve as decisive test, is the dependence of the scattering function on the momentum transfer.

To demonstrate this consequence of the Fermi statistic let us consider two-sublattice system in which intersublattice interaction dominates, i.e. \(A_{\kappa \kappa^{'}} \xi = \delta_{\kappa} T^{*}/2\) for \(\xi \neq \xi^{'}, T^{*}/2\) for \(\xi = \xi^{'}, \alpha \) and negligible for \(\xi = \xi^{'}, \beta\). (Numeric calculations

\[
FIG. 1. The energy dependence of the scattering function of the spin liquid with the bandwidth \(T^{*} = 2\) for \(T = 0.1\) (thick solid line), \(T = 0.25\) (thin solid line), \(T = 0.5\) (dashed line) and \(T = 2\) (dotted line).
\]
show that these approximations, although simplify the analytic estimations, do not influence the character of presented effects.) In the framework of this model the four-fold degeneracy of the states in each elementary cell is reduced and the set of eigenstates consists of two two-fold degenerate eigenstates with the energies ±T*/2. The chemical potential is µ = 0. Let us assume for definiteness that the basis vector ξ is parallel to z-axis and the distance between the magnetic ions is d.

To calculate the momentum dependence it is convenient to introduce the function

\[ S_{sl}^{\nu}(\kappa) = \int_{-T}^{T} d(\hbar \omega) S_{sl}(\kappa, \hbar \omega), \]

which corresponds to the integral value of the cross section with respect to the energy transfer. This function represents the total quasielastic scattering on the lowest crystal field level.

In the final expression for the SL scattering function

\[ S_{sl}^{\nu}(\kappa) = \left( \frac{B(\kappa)}{2} \right) \sum_{\alpha \beta} (\delta_{\alpha \beta} - \tilde{k}_{\alpha} \tilde{k}_{\beta}) T^{\alpha \beta} D(\kappa, T), \]

where

\[ T^{\alpha \beta} = \sum_{\sigma \sigma'} (\sigma | \hat{J}_\alpha | \sigma') (\sigma' | \hat{J}_\beta | \sigma), \]

the Fermi statistics results in the additional factor

\[ D(\kappa, T) = \frac{1}{2} \left\{ 1 - \tanh^2 \left( \frac{T^*}{T} \right) \cos(\kappa d) \right\}, \]

which is equal to unity in case of Boltzmann distribution. The factor D(\kappa, T) leads at low temperatures to the oscillation of the scattering cross section with the period \( \kappa_p = 2\pi/(d \cos(\kappa d)) \). Important property for experimental check is the dependence of the oscillations period on the momentum transfer direction.

The scattering function for polycrystalline samples \( S_{sl}^{\nu}(\kappa) \) can be calculated by integration of expression \[ \int \] over the angles. To demonstrate the consequences of Fermi statistic I separated out into the factor \( S_{sl}^{\nu}(\kappa) \) the standard scattering function for Boltzmann statistics which only source of the momentum transfer dependence is the square of formfactor \( F(\kappa) \). It is seen from the following expression

\[ S_{sl}^{\nu}(\kappa) = S^{\nu}(\kappa) \times \frac{1}{2} \left\{ 1 - \tanh^2 \left( \frac{T^*}{T} \right) \left[ \left( 1 + \frac{\eta}{2} \right) \sin(\kappa d) - \frac{3}{2} \eta \Phi(\kappa d) \right] \right\}, \]

that integration does not lead to the total suppression of oscillations. Here

\[ \Phi(\kappa d) = \frac{\sin(\kappa d) - \kappa d \cos(\kappa d)}{(\kappa d)^3}. \]

For large enough momentum transfer \( \kappa d \gg 1 \) the function \( \Phi(\kappa d) \) is much less than factor \( \sin(\kappa d)/(\kappa d) \). It is seen from the upper panel of Fig. 3 that the temperature does not significantly suppress the oscillations, at least in the mean field approximation. Even at temperature \( T \sim T^* \), which is enough to destroy SL state, the oscillations amplitude is still noticeable.

One should note that the period of driven by the Fermi statistic oscillations is determined by the distance between interacting magnetic ions \( d \) and have nothing to

\[ \eta = \frac{T_{xx} + T_{yy} - 2T_{xx}}{T_{xx} + T_{yy} + T_{xx}} \]

FIG. 2. Dependence of the ratio \( (\kappa) = S_{sl}^{\nu}(\kappa)/S_{sl}^{\nu}(\kappa) \) on the momentum transfer module \( \kappa \) for SL bandwidth \( T^* = 1 \) and \( \eta = 0 \). Upper panel: in the perfect lattice \( T = 0 \) (solid line), \( T = 0.7 \) (dashed line) and \( T = 1.0 \) (dotted line). Lower panel: at zero temperature for \( \Lambda = 1 \) (solid line), \( \Lambda = 0.6 \) (dashed line) and \( \Lambda = 0.2 \) (dotted line). Insert: Scattering function taking into account the formfactor of Ce\(^{3+} \) ion for the interionic distance \( d = 4A \).
do with the peculiarities of SL Fermi surface. The characteristic distance between the magnetic ions in the HF compounds is \( d \sim 4\AA \). Therefore, in spite scattering function decrease \( S_{sl}^{tot}(\kappa) \sim F^2(\kappa) \) it is possible to observe several oscillation periods (see insert in the upper panel of Fig. 3).

6. However, the influence of the lattice imperfection on the suppression of the oscillations is much more significant. In the simplest case, when coherency is not destroyed by the absence of one of the magnetic ions, the lattice imperfection results in different environments of basis ions. The change of environment leads to the change of the wave function of the crystal field states [14]. Different wave functions of basis ions lead, in turn, to inequality of matrix elements \( \langle \sigma \mid J^\alpha \mid \sigma' \rangle_1 \neq \langle \sigma \mid J^\alpha \mid \sigma' \rangle_2 \) for sites 1 and 2. Although expression for scattering function is derived for general case I present less cumbersome formula for the isotropic one-ion scattering case (x, y and z directions are equivalent and Cartesian indices are omitted in the following). Nonoivalence of the basis ions can be characterized by the quantity

\[
U = \frac{\sum_{\sigma \sigma'} |\langle \sigma \mid \hat{J} \mid \sigma' \rangle_1 - \langle \sigma \mid \hat{J} \mid \sigma' \rangle_2|^2}{\sum_{\sigma \sigma'} |\langle \sigma \mid \hat{J} \mid \sigma' \rangle_1 + \langle \sigma \mid \hat{J} \mid \sigma' \rangle_2|^2}.
\]

Within this simple model the influence of the lattice imperfection on the scattering function

\[
S_{sl}^{tot}(\kappa) = S^{tot}(\kappa) \frac{1}{2} \left\{ 1 - \Lambda \tanh^2 \left( \frac{T^*}{T} \right) \frac{\sin(\kappa d)}{\kappa d} \right\}
\]

is determined by the factor

\[
\Lambda = \frac{1 - U}{1 + U},
\]

which is equal to unity in the perfect lattice and tends to zero for the total lost of coherency. The lower panel of Fig. 3 demonstrates the influence of the lattice imperfection on the suppression of the oscillations.

7. In conclusion, it have to be noted that dependence of the spin liquid neutron scattering cross section on the energy transfer can not be distinguished from the scattering on the relaxing spin. However, the oscillatory dependence of the cross section on the momentum transfer is unique for spin liquid of RVB type. Besides, in case of soft enough crystal field splitting \( \Delta_{CF} \sim T^* \) one have to include into the Hamiltonian \( \parallel \) the terms which are nondiagonal with respect to crystal field levels \( \parallel \). This terms lead to the oscillations of the inelastic scattering intensity. The oscillations of the inelastic scattering intensity were experimentally observed \[15\] in Kondo-semimetal CeNiSn and quantitatively explained in the framework of spin liquid conception \[16\]. However, since many characteristics of the periodic systems demonstrate the oscillatory behavior in neutron scattering response (e.g. the peak width oscillations in paramagnets \[17\]), it is important to find the property which is connected with RVB correlations unambiguously.

As it is shown in the present paper, the oscillations of the total quasielastic scattering cross section is direct consequence of the Fermi statistic of RVB elementary excitations. Therefore, an experimental observation of the predicted effect might serve as notable argument in favor of the validity of spin liquid conception for the description of heavy fermion systems.

I am deeply grateful to P. A. Alekseev, K. A. Kikoin and V. N. Lazukov for critical discussions. This work was supported by Russian Fund for Fundamental Research (project No. 98-02-16730).

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