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Proceedings of the 21st International Conference on Low Temperature Physics
Prague, August 8-14, 1996
Part S4 - LT Properties of Solids 1: Magnetism (theory)

Elementary excitations in Kondo-systems CeNiSn and CeRhSb

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The anomalies in the low-temperature thermodynamics (heat capacity, thermal expansion) of Kondo-materials CeNiSn and CeRhSb are explained in a framework of the theory of spin liquid in a Kondo lattice which was used previously for quantitative description of inelastic magnetic neutron scattering spectra (2.5meV and 4meV excitations). These anomalies are explained by the interplay between the heavy fermions and the crystal field excitations in assumption that the crystal-field splitting δ_{CF} is less than Kondo temperature T_K. It is shown that the V-shape pseudogap appears in a spin excitation branch but the charge excitations remain gapless. The spin excitation spectrum for an orthorhombic lattice turns out to be highly anisotropic.

1. The orthorhombic compounds CeNiSn and CeRhSb, apparently, form a special group in the family of Kondo materials. These compounds demonstrated quasi semiconducting behavior of electrical resistivity at early stage of studies but the recent measurements on the samples of better quality show rather metallic then semiconducting temperature dependence of resistivity [1]. On the other hand the pseudogap behavior is distinctly seen in the specific heat, magnetic susceptibility and inelastic neutron scattering spectra [2]. To reconciliate the apparent contradictions the theory was offered [3] which connects the spin response and low-temperature thermodynamics of these materials with spin-liquid excitations of Fermi type (resonating valence bonds) which are strongly intermixed with the on-site crystal-field excitations under the condition $\Delta_{CF} < T_K$. In the present paper we calculate the energy spectrum of these mixed excitations for real symmetry of orthorhombic CeNiSn and use this spectrum in calculations of low-temperature specific heat and thermal expansion.

2. The Kondo lattice is described by the effective indirect exchange Hamiltonian of Cornut-Coqblin (CC) type which is obtained from the Anderson-lattice Hamiltonian

$$H_{sf} = \sum_{\mathbf{k}\Lambda} \epsilon_{k\Lambda} c^{\dagger}_{\mathbf{k}\Lambda} c_{\mathbf{k}\Lambda} - \sum_{\mathbf{kk'i}\Lambda\Lambda'} J^{\Lambda\Lambda'}_{\mathbf{ikk'}} f^{\dagger}_{\mathbf{i}\Lambda} f_{\mathbf{i}\Lambda'} c^{\dagger}_{\mathbf{k'}\Lambda'} c_{\mathbf{k}\Lambda}$$

written in assumption that the sf-hybridization $V^{\Lambda}_{\mathbf{ik}}$ intermixes only the atomic and the band states which belong to the same irreducible representation Λ of the crystal point group. Here $\Lambda = G, E$ corresponds to the ground and excited states of Ce(f^1) ion, $J^{\Lambda\Lambda'}_{\mathbf{ikk'}} = V^{\mathbf{i}*}_{\mathbf{k}\Lambda} V^{\mathbf{j}}_{\mathbf{k'}\Lambda'}/(\epsilon_k - E_f)$ is the indirect exchange interaction between the f-state and the band electrons. In the assumption $\sum_\Lambda f^{\dagger}_{\mathbf{i}\Lambda} f_{\mathbf{i}\Lambda} = 1$, the conduction electrons can be integrated out, and one comes to indirect RKKY interaction $H_{RKKY} = \sum_{\mathbf{ii'}\Lambda\Lambda'} I^{\Lambda\Lambda'}_{\mathbf{ii'}} f^{\dagger}_{\mathbf{i}\Lambda} f_{\mathbf{i}\Lambda'} f^{\dagger}_{\mathbf{i'}\Lambda'} f_{\mathbf{i'}\Lambda}$

Since the orthorhombic symmetry of CeNiSn lattice is due to small distortion of trigonal elementary cell, we use the CC approximation for the latter case and deal with the pair of doublets $|G\pm\rangle = a\,|\pm 1/2\rangle \pm b\,|\mp 5/2\rangle$, $|E\pm\rangle = |\pm 3/2\rangle$ and then take into account the non-CC exchange term which is induced by the orthorhombic component of the crystal field, $H'_{RKKY} = \sum_{\mathbf{ii'}E} \left(\tilde{I}^{GE}_{\mathbf{ii'}} f^{\dagger}_{\mathbf{i}G} f_{\mathbf{i}E} f^{\dagger}_{\mathbf{i'}G} f_{\mathbf{i'}G} + H.c\right)$.

The uniform spin-fermion state of RVB type is described in the mean-field approximation by the anomalous average $\Delta^G = \sum_\nu \langle f^{\dagger}_{\mathbf{i}G\nu} f_{\mathbf{i'}G\nu}\rangle$ (ν is the row of the irreducible representation). Inserting this correlator into the RKKY Hamiltonian one comes to the mean field Hamiltonian which describes the spinon excitations

$$H_{MF} = \sum_{\mathbf{i}} E_G f^{\dagger}_{\mathbf{i}G} f_{\mathbf{i}G} + \Delta_{CF} f^{\dagger}_{\mathbf{i}E} f_{\mathbf{i}E} + \sum_{\mathbf{ii'}} T^G_{\mathbf{ii'}} f^{\dagger}_{\mathbf{i}G} f_{\mathbf{i'}G} + \sum_{\mathbf{ii'}} \left[B^{GE}_{\mathbf{ii'}} f^{\dagger}_{\mathbf{i}G} f_{\mathbf{i'}E} + H.c.\right] \qquad (1)$$

Here $T^G_{\mathbf{ii'}} = \Delta^G I^G$, and $B^{GE}_{\mathbf{ii'}} = \Delta^G I^{GE}_{\mathbf{ii'}}$

The density of spin-fermion states described by (1) is shown in fig.1. This picture is obtained for

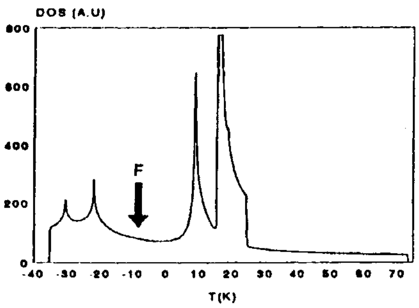

Fig. 1. Density of spin-fermion states in CeNiSn. The position of Fermi level is determined by the constraint condition.

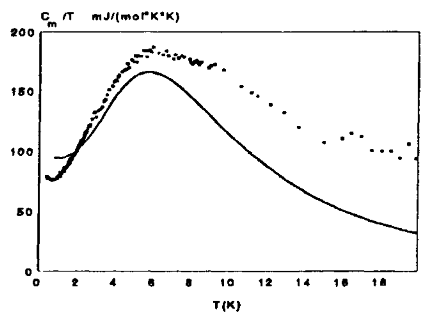

Fig. 2. C_m/T versus T for CeNiSn. Theory and experimental data are shown in line and points, resp.

the following values of parameters: $\Delta_{CF} = 15$K, $T^G_{ii'} = 11K$ for the nearest neighbors in Ce sublattice in the bc-plane of orthorhombic CeNiSn lattice, $|\mathcal{B}^{GE}_{ii'}| = 5$K for the nearest neighbors in two Ce sublattices. The assumption of 2D character of spin-fermion spectrum follows from the character of neutron scattering spectra [2]. The Kondo temperature can be estimated from this spectrum as $T_K \sim 60 - 80$K which is in reasonal agreement with experimental data [2]. $C_m(T)$ calculated in the mean-field approximation described above compared to experimental data of [4]. The spinon DOS shown in Fig. 1) results not only in low-temperature behavior $C_m(T) = aT + bT^2$ but also describes quantitatively the peak at $T \approx 6$K. According to our description this peak appears due to the influence of highest CF-induced peak in DOS.

3. It is seen that the calculated spectrum has the pseudogap structure which was postulated phenomenologically in previous studies [2] but the form of this pseudogap is determined by the van Hove singularities of spin-fermion dispersion law. The highest in energy double peak arises due to intersection

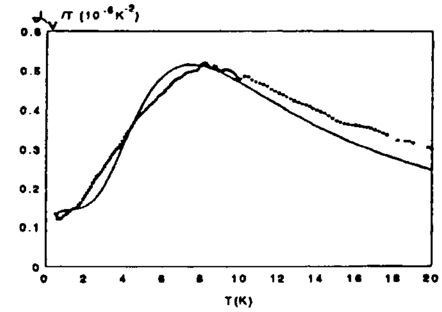

Fig. 3. α_V/T versus T for CeNiSn. Theory and experiment are shown in line and points, resp.

of CF level with spinon continuum. This DOS explains the main features (peaks at 2, 4 and 7 eV) of neutron scattering spectra and gives good quantitative description of specific heat and thermal expansion of CeNiSn at low temperatures provided the dominant contribution to the low-temperature spin entropy is ascribed to spin-fermion excitations [3]. Fig. 2 illustrates the spinon contribution to specific heat The spinon contribution to the volume expansion α_v is determined by the volume derivatives of the hybridisation integral and the CF level position, $\Gamma_B = \partial \ln \mathcal{B} / \partial \ln V$ and $\Gamma_{\Delta_{CF}} = \partial \ln \Delta_{CF} / \partial \ln V$ respectively. Then,

$$\alpha_v = k_T S(T) \left[1 + \Gamma_B \left(\frac{\partial S}{\partial B} \right)_T + \Gamma_{\Delta_{CF}} \left(\frac{\partial S}{\partial \Delta_{CF}} \right)_T \right]$$

Here k_T, S and V are the isothermal compressibility, the entrophy and the volume, resp. Using this expression with parameters $k_T = 0.25 * 10^{-11}\text{Mbar}^{-1}$, $\Gamma_B = 4.0$ and $\Gamma_{\Delta_{CF}} = 2.5$ one gets quantitative description of experimental data (fig. 3). The same theory gives the quantitative description of low-T thermodynamics of CeRhSb with tripled energy scale.

This work was supported by the grants INTAS-93-2834, NWO-07-30-002 and RFBR-95-02-04250a.

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