Nature of the pseudogap in the energy spectrum of CeNiSn

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It is shown that interaction with crystal-field excitations can cause a pseudogap to appear in the spectrum of heavy-fermion compounds with a nearly integer valence. The necessary conditions for the appearance of a pseudogap are formulated and it is demonstrated that these conditions are realized in the compound CeNiSn. Suppression of the pseudogap by an external magnetic field is studied.

1. Interest in rare-earth compounds with low carrier density has increased sharply in the last few years in view of the discovery of an entire series of Ce and U-based systems which have a gap or pseudogap in the spectrum of low-energy Fermi excitations. In the literature these compounds are called Kondo insulators.[1,2] They differ in many respects from the well-known semiconductor compounds with intermediate valence and a microgap of several meV in the spectrum and they are based on elements from the center or end of the $4f$ series—SmS, SmSe, SmB_6, TmTe, and YbB_{12}. Among these systems, which have very diverse properties, the equiatomic triple compounds CeNiSn (Refs. 3 and 4) and CeRhSb, (Ref. 5), whose spectra contain energy gaps (or pseudogaps) with widths $\Delta \approx 3$ K and 4 K, respectively, form a special group. These gap widths appear to be anomalously small, even compared with the values of Δ presented above for "classical" semiconductors with intermediate valence. The existence of a pseudogap in the spectrum of Fermi excitations is manifested in the thermodynamic, kinetic, and relaxational properties. In addition, these properties are extremely sensitive to an external magnetic field (see, for example, Ref. 6), and the response to such a field depends on the orientation of the field with respect to the crystalline axes of the orthorhombic lattice, in which these compounds crystallize.

In this letter we propose a model that explains the nature of low-energy Fermi excitations in these systems, the reason for the appearance of a pseudogap in the spectrum, and the suppression of the pseudogap by an external magnetic field. The theory is based on the assumption stated in Ref. 7 about the spin nature of the Fermi excitations (heavy fermions) in compounds with nearly integer valence, and it gives a qualitative explanation of the main physical properties of CeNiSn.

2. A model describing systems with heavy fermions (HFs) and integer valence was formulated in Refs. 7 and 8. This model describes systems such as strongly correlated electronic liquids, whose low-temperature thermodynamics is determined predominantly by spin excitations which obey Fermi statistics in the limit $T \to 0$, lie in a narrow energy band with energy of the order of T^*, and do not carry charge. These

excitations interact with the charged Fermi liquid formed by the conduction electrons and renormalize the spectrum of the liquid, thereby influencing the physical properties associated with charge transfer. Here T^* is the temperature at which collective interactions in the spin subsystem are destroyed. Excitations of one other type, which do not carry charge, are also observed in such HF systems: transitions between crystal-field-split f levels of the rare-earth element. The energy of these excitations is characterized by the parameter Δ_{CEF}, which generally exceeds the spin temperature T^*, so that the crystal field has virtually no effect on the spectrum of the HFs at low temperatures, $T < T^*$. In this letter we show that in the limit of weak splitting,

$$\Delta_{CEF} < T^*, \tag{1}$$

the excitations of the crystal-field levels, as a result of interacting with heavy spin fermions, can significantly restructure the spectrum of the fermions and account for the formation of an energy gap or a pseudogap in the spectrum in crystals with lower than cubic symmetry.

We will use the conventional Coqblin–Schrieffer approximation for an Anderson lattice to describe HF systems under conditions where the inequality (1) is satisfied. In this approximation the states of the conduction electrons are expanded in partial amplitudes, and the hybridization integrals of these electrons with the atomic states do not mix different partial waves. The Hamiltonian of the Anderson lattice in this approximation has the form

$$H = H_f + H_{\text{band}} + H_{\text{hyb}} + H'. \tag{2}$$

Here

$$H_f = \sum_{in} \sum_{\Gamma} E_{f\Gamma}^{(n)} |in\Gamma)(in\Gamma|. \tag{3}$$

This term in the Hamiltonian describes the sublattice of f-ions in the configuration f^n. In the case of the ion $Ce^{3+}(f^1)$ the quantum numbers $\Gamma = JM$—the total angular momentum (J) and its projection (M)—assume the values $J = 5/2$ ($5/2 \geqslant M \geqslant -5/2$). The energy $E_{f\Gamma}^{(n)}$ is the energy level of the f^n ion. The band Hamiltonian

$$H_{\text{band}} = \sum_{\mathbf{k}} \sum_{\gamma} \epsilon_{\kappa} b_{k,\gamma}^{+} b_{k,\gamma} \tag{4}$$

describes conduction electrons characterized by the wave vector $\mathbf{k}$ for the partial wave $\gamma = jm$. The third term

$$H_{\text{hyb}} = \sum_{\mathbf{ki}} \sum_{\gamma,\Gamma,} V_{\mathbf{ki}}^{\gamma} b_{\mathbf{k}\gamma}^{+} |\mathbf{i},0)(in\Gamma| + \text{H.c.} \tag{5}$$

describes hybridization of an f electron, at the site i, with a conduction electron. As a result, the configuration of the cerium ion changes from f_{Γ}^{1} to f^0. In the Coqblin–Schrieffer approximation we have $\gamma = \Gamma$. The last term H' contains all interactions which are outside the range of the Coqblin–Schrieffer approximation.

In order to allow for the splitting of the f levels in a crystal field we must switch from the angular momentum representation to the irreducible representations of the

point group of the crystal: $(\Gamma,\gamma)\to(\Lambda,\gamma)$. In noncubic crystals the multiplet Γ of the configuration $f^1_{5/2}$ splits into three Kramers doublets. We present below the wave functions which describe the states Λ of these doublets for the three most often encountered types of crystal symmetry in HF compounds:

hexagonal crystal field

$$|1\pm\rangle=|\pm 5/2\rangle,\quad |2\pm\rangle=|\pm 3/2\rangle,\quad |3\pm\rangle=|\pm 1/2\rangle; \tag{6}$$

tetragonal crystal field

$$|1\pm\rangle=a|\pm 3/2\rangle+b|\mp 5/2\rangle,\quad |2\pm\rangle=|\pm 1/2\rangle,$$

$$|3\pm\rangle=a|\mp 5/2\rangle-b|\pm 3/2\rangle; \tag{7}$$

trigonal crystal field

$$|1\pm\rangle=a|\pm 1/2\rangle+b|\mp 5/2\rangle,\quad |2\pm\rangle=\pm 3/2\rangle,$$

$$|3\pm\rangle=a|\mp 5/2\rangle-b|\pm 1/2\rangle. \tag{8}$$

It is obvious that these states hybridize differently with different Bloch waves. For example, waves with $m=\pm 1/2$ hybridize with the state $|3\rangle$ of the hexagonal multiplet of the crystal field, with the state $|2\rangle$ of the tetragonal multiplet, and with the states $|1\rangle$ and $|3\rangle$ of the trigonal multiplet. The waves with $m=\pm 3/2$ in turn hybridize with the states $|2\rangle$ of the hexagonal and trigonal multiples and with the states $|1\rangle$ and $|3\rangle$ of the tetragonal multiplet. Finally, the waves with $m=\pm 5/2$ mix only with the state $|1\rangle$ of the hexagonal multiplet and with the states $|1\rangle$ and $|3\rangle$ of the tetragonal and trigonal multiplets. Thus, in the presence of an interaction with any partial Bloch wave some levels from the crystal-field multiplet remain unhybridized. Since the hybridization interaction is primarily responsible for the formation of the HF state in an Anderson lattice, we conclude that if only a partial Bloch wave makes the dominant contribution to hybridization at the Fermi level, some states from the crystal-field multiplet do not participate in the formation of the coherent HF state. As will be shown below, however, these levels can significantly influence the low-energy part of the spectrum of elementary excitations if the condition (1) is satisfied.

Let us assume for simplicity that only one of the Kramers doublets (6)–(8) participates in the formation of an HF band in the energy interval $0<E<T^*$. We designate it as $|a\rangle$. Next, let $|\beta\rangle$ be the lowest of the doublets which remains unhybridized with the wave which dominates at the Fermi surface. In accordance with the condition (1), we assume that its energy $E_\beta<T^*$. We assume that the third level of the crystal field has a higher energy which we will not consider here. The unhybridized part of the atomic Hamiltonian H_f can then be re-expanded in terms of the crystal-field states $|i\Lambda\rangle$ and we can switch to the pseudofermion representation $|i\Lambda\rangle\langle i\Lambda|=f^+_{i,\lambda}f_{i,\lambda}$, $\Lambda=\alpha,\beta$, as is usually done in the theory of the Anderson lattice with infinite Hubbard repulsion U and nearly integer valence (see, for example, Refs. 9 and 10). As a result, the atomic part of the Hamiltonian assumes the form

$$H=H_\alpha+H_\beta, \tag{9}$$

where

$$H_{\Lambda=\alpha,\beta,}=\sum_{i}\epsilon_{\Lambda}f_{i\Lambda}^{+}f_{i\Lambda}. \tag{10}$$

The conventional local constraint

$$\sum_{\Lambda}f_{i\Lambda}^{+}f_{i\Lambda}=1-\delta_{i} \tag{11}$$

is imposed on the system. Here $\delta\ll 1$ is the deviation of the valence of the $f$ion from an integer. By definition, a single partial Bloch wave $|c\mathbf{k}\alpha)$ predominates in the hybridization near the Fermi surface. The spin excitations, which are described by a part of the Hamiltonian $H_{\alpha}+H_{\text{band},\alpha}+H_{\text{hyb},\alpha,}$, can then be projected, with the help of one of the well-known methods of the theory of heavy fermions, onto a narrow energy interval $\sim T^{*}$ near the Fermi level, which plays the role of the origin of the energy scale. The choice of projection method is not important in what follows, since all known procedures[9-13] ultimately lead to the formation of a coherent band of Fermi excitations predominantly of a spin nature,[7] $|i\alpha\sigma)\rightarrow|\mathbf{k}\tilde{\alpha}\sigma)$ with a dispersion $\epsilon_{\mathbf{k}\alpha}$ and band width $\sim T^{*}$. Next, we see that the remaining part H_{β} of the atomic Hamiltonian (9) can be regarded as a dispersion-free pseudofermion level which is in "resonance" with the spin-fermion continuum by virtue of the inequality (1). However, the weak perturbation associated with the term H' in the Hamiltonian (2) destroys the crystal-field classification (6)–(8), which ultimately results in mixing of the states $|\mathbf{k}\tilde{\alpha})$ and $|\beta)$. As a result, we obtain an effective Hamiltonian for low-lying Fermi-type spin excitations:

$$H=\sum_{\mathbf{k}\sigma}\epsilon_{\mathbf{k}\alpha}\tilde{f}_{\mathbf{k}\alpha\sigma}^{+}\tilde{f}_{\mathbf{k}\alpha\sigma}+E_{\beta}\sum_{\mathbf{k}\sigma}f_{\mathbf{k}\beta\sigma}^{+}f_{\mathbf{k}\beta\sigma}+\sum_{\mathbf{k}\sigma}(g_{\mathbf{k}}^{\alpha\beta}\tilde{f}_{\mathbf{k}\alpha\sigma}^{+}f_{\mathbf{k}\beta\sigma}+\text{H.c.}). \tag{12}$$

Here $g_{\mathbf{k}}^{\alpha\beta}$ is the above-mentioned secondary-hybridization parameter, the operators $\tilde{f}_{\mathbf{k}\alpha\sigma}^{+}$ describe heavy fermions, and the operators $f_{\mathbf{k}\beta\sigma}$ are the Fourier transforms of the pseudofermion operators $f_{\beta,\sigma}$, which describe a Kramers doublet in the Hamiltonian H_{β} (10). We replace the local constraint (11) for spin-fermions by the global constraint (compare Ref. 13)

$$\sum_{\mathbf{k}\sigma}[\langle f_{\mathbf{k}\alpha\sigma}^{+}f_{\mathbf{k}\alpha\sigma}\rangle+\langle f_{\mathbf{k}\beta\sigma}^{+}f_{\mathbf{k}\beta\sigma}\rangle]=1-\delta. \tag{13}$$

This condition is introduced into the effective Hamiltonian with the help of a Lagrange multiplier μ, which plays the role of a chemical potential for fermion occupation numbers.

In order to estimate the intermultiplet hybridization parameter $g_{\mathbf{k}}^{\alpha\beta}$, we introduce a simple perturbation mechanism, which is associated with the existence in the Hamiltonian H' of terms proportional to the integral $(c\mathbf{k}\alpha|V'|\beta)$, where the low-symmetric part of the crystal field V' is responsible for the mixing partial waves. We then obtain the following expression for $g_{\mathbf{k}}^{\alpha\beta}$.

$$g_{\mathbf{k}}^{\alpha\beta}=\frac{(\tilde{f}\mathbf{k}\alpha|V|c\mathbf{k}\alpha)(c\mathbf{k}\alpha|V'|f\mathbf{k}\beta)}{D}, \tag{14}$$

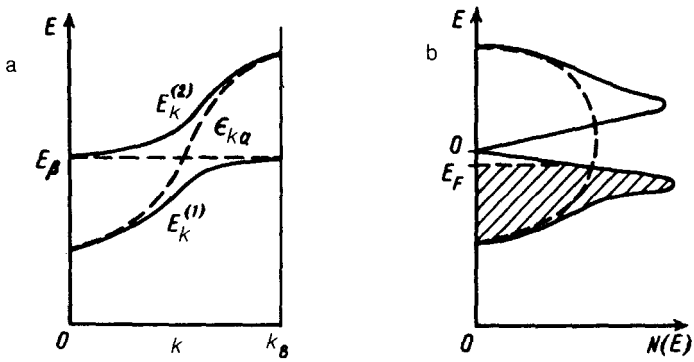

FIG. 1. a) Two-band spectrum for spin-fermion excitations and b) density of states $\mathcal{N}(E)$ and the Fermi level E_f determined by the condition (13).

where D is the width of the conduction band. Estimate of this expression gives $g_{\mathbf{k}} \simeq \xi T^*$, where $\xi = \bar{V}'/\bar{V} \ll 1$. Here $\bar{V}'$ and $\bar{V}$ are the values of the hybridization integrals between the f electrons and their nearest neighbors in the corresponding sublattices in the site representation.

The energy spectrum of the spin-fermion excitations is described by the equation

$$E_{\mathbf{k}\sigma}^{(1,2)} = \tfrac{1}{2}[(E_\beta + \epsilon_{\mathbf{k}\alpha}) \pm \sqrt{(E_\beta - \epsilon_{\mathbf{k}\alpha})^2 + |g_{\mathbf{k}}|^2}]. \tag{15}$$

This spectrum and the corresponding density of states are shown schematically in Fig. 1. Because the hybridization matrix elements are not confined in space, the integral $g_{\mathbf{k}}^{\alpha\beta}$ has nodes at the high-symmetry points of the Brillouin zone, and a pseudogap appears in the spectrum of elementary excitations. It is easy to see that when the level E_β is situated approximately at the center of the HF band, the Fermi level for spin-fermions, which is determined by the condition (13), is situated in the pseudogap, where the density of states $\mathcal{N}(E)$ is sensitive to the energy difference $E - E_F$, so that the low-temperature thermodynamics of the system is determined by the structure of the spectrum in the region of this pseudogap. The form of $\mathcal{N}(E)$ depends on the type of lattice, but for small $\epsilon = E - E_\beta$ it is assumed to be a linear function of ϵ.

3. We now consider the effect of an external magnetic field on the spectrum of spin-fermion excitations. If the magnetic field is applied parallel to the quantization axis of the magnetic moment ($h = h_\parallel$), the Zeeman term has the form $H_{\text{Zeeman}} = h \sum_i J_i^z$. This term splits the level $E_{\beta\sigma}(h) = E_\beta + \sigma \mu_B g_\beta h$, where g_β is the effective g-factor of the given Kramers doublet, $\sigma = \pm 1/2$. The effect of the magnetic field on the heavy-fermion states is more complicated, since in the Ce-based systems the single-site (Kondo) correlations and the interstitial correlations (those associated with indirect exchange) affect the low-energy excitations (see, for example, the experimental study[14] of the properties of $CeRu_2Si_2$). We restrict the discussion to weak fields, which do not suppress magnetic correlations but only orient the spins. The spectrum of heavy fermions in an external field will then have the form $\epsilon_{k\alpha\sigma} = \epsilon_{k\alpha} + 2\mu_B \sigma h (\sigma = \pm 1/2)$. It is easy to see that for $g_\beta > 2$, which holds for all Kramers doublets, with the

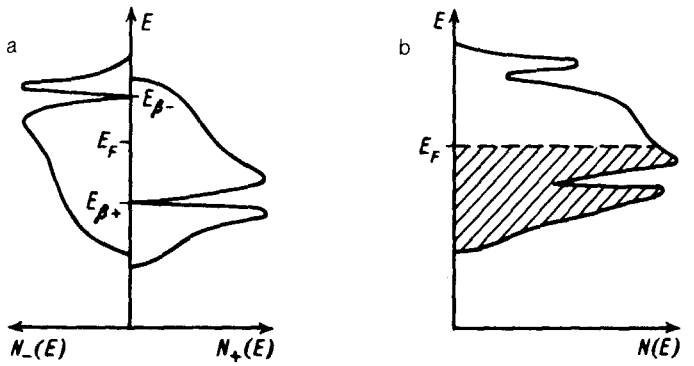

FIG. 2. a) Density of states $\mathcal{N}_\sigma(E)$ for projections $\sigma = \pm 1/2$ for $h > h_\parallel^{cr}$ and b) total density of states $\mathcal{N}(E)$ for $h > h_\parallel^{cr}$.

exception of the states $|3)$ in (6) and $|2)$ in (7), there exists a critical field $h_\parallel^{cr} \sim \xi T^*$, corresponding to detuning of at least one of the spin sublevels $E_{\beta\sigma}$ from resonance with the Fermi level (Fig. 2). For $h > h_\parallel^{cr}$ the "standard" behavior of the HF system is thus restored.

Assume now that the field is applied perpendicular to the quantization axis, which is determined by the magnetic anisotropy energy $-D\Sigma_i(J_i^z)^2$, for example, in the x direction. The Zeeman term in the Hamiltonian will then have the form $H_{Zeeman} = \frac{1}{2}h_\perp (J^+ + J^-)$. The operators $J^\pm$ do not have diagonal elements in the pseudofermion representation, so that the only effect of the magnetic field is an additional contribution to the effective hybridization $g^{\alpha\beta}$. For this reason the transverse field $h_\perp$ cannot have an appreciable effect on the gap in the HF spectrum.

Thus we see that the proposed model not only explains the appearance of a pseudogap in the spectrum of elementary excitations of a heavy-fermion system, but it also describes the anisotropy of the systems response to an external magnetic field. It should be noted, however, that this spectrum describes only spin-origin Fermi excitations.[7] The influence of the gap in the spin spectrum on charge-type Fermi excitations (conduction electrons) requires a separate investigation. Leaving this investigation for a future work, here we analyze the applicability of the proposed model to the case of the compound CeNiSn and we consider briefly its low-temperature thermodynamics and kinetics.

4. Having obtained a general picture of the pseudogap in the spectrum of Fermi excitations in the spin subsystem of a Kondo lattice, we must now determine the applicability of this picture to real compounds of the type CeNiSn. First, we note that this system satisfies the main necessary conditions for the appearance of a gap.

a) The crystalline environment of the Ce ion in the orthorhombic lattice of CeNiSn is a slightly distorted hexagonal prism formed by alternating Ni and Sn atoms,[15] i.e., the crystal field has mainly trigonal symmetry with weak orthorhombic perturbation. Thus we can construct a description of spin excitations in the basis (8),

taking as the perturbation V' in Eq. (14) the orthorhombic component of the crystal field.

b) Measurement of the photoemission spectra of CeNiSn has shown[16] that, first, the valence of Ce in this compound is close to $+3$ and, second, the Fermi edge of this spectrum is formed predominantly by the $5p$ electrons of the Sn ions, so that the wave $|\mathbf{k}\,\frac{3}{2}m)$ can be chosen as the predominant Bloch component in the Coqblin–Schrieffer Hamiltonian, and then the doublet $|1\sigma)$ from the multiplet (8) will play the role of the resonance state $|\beta)$ in Eq. (12).

c) Splitting of the crystal-field levels in this system was not observed, in agreement with the theory. However, in the compound closest in composition—the tetragonal crystal $CeNi_2Sn_2$—Δ_{CEF} was found to be very small (≈ 17 K) and close to $T^* \approx 7$ K.[17] It can thus be assumed that in orthorhombic coordination the crystal field of Sn and Ni at the Ce sites is weak, and the main assumption of the theory (1) is satisfied for CeNiSn.

Thus all the necessary prerequisites for a pseudogap to appear in the spectrum of spin-fermion excitations in CeNiSn are present. The proposed model explains the anomalous thermodynamic properties of this compound. It has been pointed out that the spectrum of Fermi excitations with density of states, near the Fermi level, $\mathcal{N}(\epsilon) \sim \epsilon$ of the type displayed in Fig. 1 explains the behavior of the heat capacity (quadratic term in the temperature dependence + a linear term) and the static magnetic susceptibility (the temperature dependence $1/T_1 \sim T^3$ for the relaxation rate of the NMR spectra). In this model the Fermi excitations are of spin origin, but the temperature dependence of the heat capacity and susceptibility remains the same as in the case of a charged Fermi liquid.

We also note that the anisotropic pseudogap in the spectrum of spin excitations has been observed directly in an inelastic neutron scattering experiment.[19]

The model also explains the reaction of the system to an external magnetic field. It is well known that a field applied parallel to the a axis of a CeNiSn crystal suppresses the gap in the excitation spectrum and restores the "normal" heavy-fermion behavior with the Sommerfeld constant $\gamma \approx 120$ mJ/K$^2 \cdot$ mole at $h = 12$ T.[6] At the same time, the pseudogap is virtually unaffected by a field oriented along the other two axes. The a axis is the quantization axis of the magnetic moment in CeNiSn, since the Ce ions are aligned in zig-zag-like chains parallel to this axis, so that $h \parallel a$ results in the effect illustrated in Fig. 2. Such restructuring of the spin-excitation spectrum by an external field also influences other properties of the system. It was shown above that the spin-fermion spectrum of CeNiSn is formed by mixing of two branches of spin excitations—"normal" HFs, which are strongly bound magnetic states with a binding energy $\sim T^*$, and crystal-field excitations, which are assumed to be almost free from the standpoint of the moment reorientation energy. The character of the magnetic response should therefore change upon transition from a case in which the magnetic Fermi surface is formed mainly by weakly bound spin-fermions (Fig. 1) to the case in which it is formed by normal HFs. This means that, in particular, a unique metamagnetic transition should occur in the system at $h_\parallel \approx h_\parallel^{cr}$. Such a transition has indeed been observed experimentally.[20]

In a magnetic field $h_{\|} > h_{\|}^{cr}$ at $T < 6$ K the temperature dependence of the electric resistance changes. The anomalous behavior with a negative temperature coefficient is replaced by the typical temperature behavior for fermion-fermion scattering, $\rho(T) \sim T^2$ (Ref. 6), at temperatures $T < 4$ K. Additional studies of the mechanisms of electron scattering by spin excitations with a pseudogap spectrum are required in order to describe the unusual low-temperature behavior of $\rho(T)$ in the absence of a field. The model explains in a natural manner, however, the fact that in strong fields the situation typical of HF systems is restored. For $h_{\|} > h_{\|}^{cr}$, when the density of HF states has the form shown in Fig. 2b, the scattering of conduction electrons by spin-origin HFs should give a quadratic behavior of $\rho(T)$ at low temperatures,[7] $T \ll T^*$.

In summary, the main properties of the compound CeNiSn can be explained by a single approach based on the assumption that in compounds with nearly integer valance the heavy fermions are of spin origin.

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