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Kondo lattice without Nozières exhaustion effect

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Abstract. – We discuss the properties of layered Anderson/Kondo lattices with metallic electrons confined in 2D xy planes and local spins in insulating layers forming chains in the z direction. Each spin in this model possesses its own 2D Kondo cloud, so that the Nozières' exhaustion problem does not occur. The high-temperature perturbational description is matched to exact low-T Bethe-ansatz solution. The excitation spectrum of the model is gapless both in charge and spin sectors. The disordered phases and possible experimental realizations of the model are briefly discussed.

The famous exhaustion problem formulated by Nozières [1] states that the number of electrons eligible to participate in Kondo screening is not enough to screen magnetic moments localized in each site of periodic Anderson lattice (AL) or Kondo lattice (KL). In spite of his latest revision [2] based on mean-field $1/N$ expansion, this exhaustion is a stumbling stone on the way from exactly solvable Anderson or Kondo impurity model [3] to the 3D AL/KL models, which are believed to be the generic models for heavy-fermion materials [4]. The problem arises already for concentrated Kondo alloys, where the number or localized spins N_i is comparable with the number of sites $N = L^n$ in the n-dimensional lattice. In this case the number of spin degrees of freedom provided by conduction electrons in a KL is not enough for screening N_i localized spins. As an option a scenario of dynamical screening was proposed [5,6], where only part of spins screened by Kondo clouds form magnetically inert singlets. The low-temperature state of such KL is a quantum liquid, where N_s singlets are mixed with $N - N_s$ "bachelor" spins, which hop around and exchange with singlets thereby behaving as effective fermions. Nozières' exhaustion is measured by a parameter $p_N = N_i/(\rho_0 T_K)$ (the number of spins per screening electron). Here $T_K \sim \rho_0^{-1} \exp[-1/(\rho_0 J)]$ is the energy scale of Kondo effect, J is the exchange coupling constant in the single-impurity Kondo Hamiltonian, ρ_0 is the density of states on the Fermi level of metallic reservoir.

Second obstacle, which does not allow the extrapolation of Kondo impurity scenario to KL is the indirect RKKY exchange $I_{jj'}$ between the localized spins, which arises in the 2nd order in J or in the 4th order in V (hybridization parameter in the generic AL Hamiltonian). The corresponding energy scale is

$$I = J^2 \chi^c_{jj'} \sim \rho_0 J^2, \tag{1}$$

where $\chi^c_{jj'} = N^{-1}\sum_{\boldsymbol{q}} \chi_c(\boldsymbol{q}) \exp[i\boldsymbol{q}\cdot\boldsymbol{R}_{jj'}]$ and $\chi_c(\boldsymbol{q})$ is the spin susceptibility of the electron gas. The Fourier transform of $\chi_c(\boldsymbol{q})$ is an oscillating function, which strongly depends on the distance $R_{jj'}$. If $I < 0$ at an average inter-impurity distance, and $|I| \sim T_K$, then the trend to inter-site antiferromagnetic coupling competes with the trend to the one-site Kondo singlet formation (Doniach's dichotomy [7]).

This competition prompted several possibilities to bypass the exhaustion limitations. According to a scenario offered in [8,9], in the critical region $|I| \sim T_K$ of Doniach's phase diagram, where the magnetic correlations are nearly suppressed by the on-site Kondo coupling, the spin liquid phase enters the game. This phase is characterized by the energy scale

$$\mathcal{I}(T) = J^2\chi^s_{jj'}(T), \tag{2}$$

where $\chi^s_{jj'}$ is the spinon susceptibility and $\mathcal{I}(T)$ is renormalized due to Kondo processes exchange integral. The condition $\mathcal{I}(T_K)>T_K$ is easily achieved both in the 3D and 2D case. The Kondo screening is then quenched in the weak-coupling regime at $T > T_K$, so that the spin degrees of freedom remain decoupled from the electron Fermi-liquid excitations both at high temperatures $T \gg T_K$ and at low temperatures $T \ll T_K$ (Curie and Pauli limit for magnetic response, respectively). At $T\to 0$ the KL behaves as a two-component Fermi liquid with strongly interacting charged electrons and neutral spinons [10]. This scenario develops on the background of strong AF correlation. It includes the possibility of ordered magnetic phases with nearly screened magnetic moments and, in particular, the quantum phase transitions. Due to separation of spin and electron degrees of freedom, Luttinger's theorem in its conventional Fermi-liquid form is invalid in this state: f-electrons represented by their spin degrees of freedom give no contribution in the formation of the electron Fermi surface. Such state is referred as a "small Fermi surface regime" in the current literature.

Another scenario for small Fermi surface regime was proposed in [11]. This scenario appeals to systems where the magnetic order is either fragile or entirely absent due to magnetic frustrations (*e.g.*, to triangular lattices). A spinon gap carrying unit flux of Z_2 gauge field is expected to arise in spin subsystem, and this gap prevents formation of Kondo singlets for a finite range of T_K. As a result, Nozières' exhaustion does not occur, and fractionalization of excitations into spin-fermions and electrons exists as in the previous case. A possibility of forming the spin liquid with $U(1)$ gauge group and spin density wave ground state has also been pointed out in [11].

In the present paper, we propose an alternative paradigm for fermion fractionalization in Kondo lattices, which possesses the generic properties of KL but is not subject to the exhaustion limitations. Namely, we consider the structures, where the number of reservoirs for Kondo screening is the same as the number of spins in a KL. This paradigm may be realized in strongly anisotropic Kondo lattices, where the metallic electrons are confined in 2D planes interlaid by insulating layers containing magnetic ions. Then the 3D reservoir of screening electrons is defragmented into L planar reservoirs. Each plane still possesses the macroscopic number of spin degrees of freedom $\sim L^2$ enough for Kondo screening, *provided* the concentration of magnetic centers per metallic plane remains small. The spin liquid features may be observed in these systems if the distribution of magnetic centers is also anisotropic, namely, if they form chains oriented in the z direction, and the inter-chain interaction is negligibly small.

We postpone the discussion of experimental realization of such systems to the concluding section and begin with the theoretical description of an ideal configuration, where all chains penetrate the stack in z direction (fig. 1a). The AL Hamiltonian for the quasiperiodic model of conduction electrons confined in metallic layers (xy plane), and magnetic ions localized in

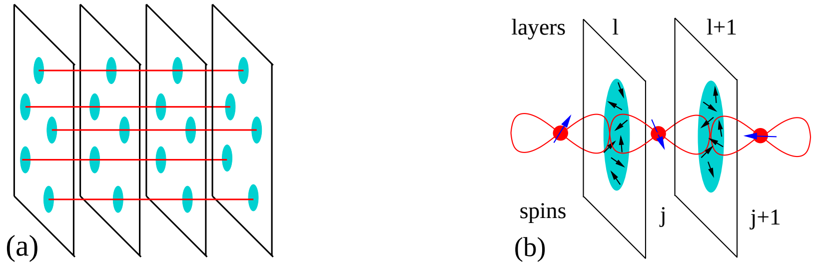

Fig. 1 – (a) Layered lattice of spatially separated charges in planes and spins in chains. (b) A fragment of a chain with Kondo clouds formed as "shadows" in metallic layers.

insulating layers between metallic planes is

$$H = \sum_{l\mathbf{k}\sigma} \epsilon_k c^\dagger_{l\mathbf{k}\sigma} c_{l\mathbf{k}\sigma} + \sum_{j\sigma}\left(\epsilon_d n^d_{j\sigma} + \frac{1}{2} U n^d_{j\sigma} n^d_{j\bar{\sigma}}\right) + + \sum_{jl}\sum_{\boldsymbol{k}\sigma}\left(V_{\boldsymbol{k}} c^\dagger_{l+1\boldsymbol{k}\sigma}(d_{j\sigma} + d_{j+1,\sigma}) + \text{H.c.}\right). \quad (3)$$

Here $\boldsymbol{k}$ is a 2D wave vector, the discrete indices numerate metallic layers l with a lattice constant $a_\parallel$ and magnetic sites j along the chains with a spacing a_z. The coupling constant $V_{\boldsymbol{k}}$ characterizes hybridization between itinerant 2D electrons in a plane $l+1$ and localized states in two adjacent sites j, and $j+1$ of the chain (fig. 1b). We treat the electrons in metallic planes in terms of Bloch waves $c_{l\boldsymbol{k}\sigma}$, while the localized electrons are characterized by Wannier functions $d_{j\sigma}$. The periodicity of magnetic sites in the xy plane is not demanded, but the average distance λ between the impurities within a layer exceeds the radius of Kondo cloud, *i.e.* satisfies the condition $\lambda \gg \hbar v_F/T_K$ (v_F and T_K are Fermi velocity of 2D electrons and Kondo temperature, respectively). There is no interaction between the chains under this condition, and a single chain represents the z component of excitation spectrum. Besides, all chains contribute to the xy-component of the spin and charge response of the AL. The effects associated with the inter-chain exchange will be discussed in the concluding part.

We came to a situation where L two-dimensional Fermi reservoirs, each with capacity L^2, screen N_i magnetic moments arranged in such a way that the effective concentration of these moments per metallic layer is $n_i = N_i/L^2$ and satisfies the condition $n_i a_\parallel^2 \ll 1$. This capacity is enough to form a screening Kondo cloud for each magnetic site within a given layer $l+1$ independently of all other sites belonging to the same layer. On the other hand, two magnetic ions localized one above another in neighboring insulating layers j, $j+1$ share the same metallic screen (see fig. 1b). Replicating these dimers along the z-axis, one comes to a system of spin chains, interacting with a system of metallic layers stacked up in the xy plane. Elimination of the hybridization term V in the Hamiltonian (3) in accordance with the standard Schrieffer-Wolff procedure, results in effective exchange Hamiltonian for each chain,

$$H^{cd}_{int} = \sum_{m=1,k,k'}^{N_i} J_{\boldsymbol{k}\boldsymbol{k}'} \vec{s}^{\,m}_{\boldsymbol{k}\boldsymbol{k}'} \vec{S}_m \quad (4)$$

(see fig. 1b with $2m \to j$, $2m-1 \to l$). Here we use notations: $\vec{S}_m = \frac{1}{2} d^\dagger_{2m,\sigma}\ \vec{\sigma}_{\sigma\sigma'} d_{2m,\sigma'}$, $\vec{s}^{\,m}_{\boldsymbol{k}\boldsymbol{k}'} = \frac{1}{2}[c^\dagger_{2m-1,\boldsymbol{k}\sigma} + c^\dagger_{2m+1,\boldsymbol{k}\sigma}]\vec{\sigma}_{\sigma\sigma'}[c_{2m-1,\boldsymbol{k}'\sigma'} + c_{2m+1,\boldsymbol{k}'\sigma'}]$. The exchange integral is $J_{\boldsymbol{k}\boldsymbol{k}'} \sim V^*_{\boldsymbol{k}} V_{\boldsymbol{k}'}/U$. We assumed the periodic boundary conditions, namely $\vec{S}_N = \vec{S}_1$ and $\vec{s}_N = \vec{s}_1$, which imply equal number of spins and planes. The open and twisted boundary conditions may be also imposed. Subtle effects modifying the ground state in these cases will be considered elsewhere.

Thus, the original model is reduced to the anisotropic KL formed by the system of 1D spin chains penetrating the stack of 2D metallic layers. Each spin creates two Kondo clouds in adjacent planes, and two neighboring spins see each other through a metallic screen by means of indirect RKKY-like exchange. This exchange may be either ferromagnetic (FM) or antiferromagnetic (AFM). The latter case is considered in terms of Doniach's dichotomy [7]. In our model this dichotomy should be reformulated. Since the long-range AFM ordering is impossible in 1D chain, two competing phases are Kondo singlet and spin liquid. Complete Kondo screening is not forbidden by Nozières' exhaustion principle, since the 2D screening layer is available for each spin in the chain. The Kondo screening is characterized by the energy scale T_K. Thus, the competing phases in the anisotropic KL are the Kondo singlet phase and the homogeneous spin liquid of RVB type with the energy scale given by eq. (2).

In order to describe the Doniach-like phase diagram we adopt the method of [9]. Namely, we derive an effective action functional by integrating out all "fast" fermionic degrees of freedom with the energies $\sim D_0$, where $2D_0$ is the conduction bandwidth. The "slow" modes give us a hydrodynamic action. Due to strong quasi-1D anisotropy there is no need in appealing to the mean-field approximation. After elimination of conduction electrons with the energies $D_0 > \varepsilon > T$ in metallic layers, the coupling J is enhanced, $J \to \tilde{J} = 1/(\rho_0 \ln(T/T_K))$ and the indirect RKKY-like spin-spin interaction mediated by the in-plane electrons [12] arises along the chains:

$$H_{int}^{dd} = -I \sum_{j,\sigma\sigma'} d_{j\sigma}^{\dagger} d_{j+1,\sigma} d_{j+1,\sigma'}^{\dagger} d_{j\sigma'}. \tag{5}$$

Here I is defined in eq. (1) with $\chi_{j,j+1}^{c}(R) = N^{-1} \sum_{\boldsymbol{q}_\parallel} \chi_c(\boldsymbol{q}_\parallel) \exp i(\boldsymbol{q}_\parallel \boldsymbol{R})$, R characterizes the relative distance between two spin projections on a plane. If $R = 0$, the chains are straight and the interaction between spins is ferromagnetic. A model of single spin chain penetrating the stack of 2D metallic planes may be mapped on that of FM spin chain interacting with an array of 1D metallic wires [13]. The Kondo screening develops similarly to a two-site Kondo model [14]. Behavior of dilute system of FM coupled spin chains interacting with arrays of 1D fermions deviates from the two-site Kondo scenario. It will be discussed elsewhere. For $R \sim a_\parallel$ the chains have a zigzag shape, with AFM interaction. Leaving the FM case for further studies, we concentrate here on the array of AFM coupled chains. Since $V/U \ll 1$, we adopt the nearest-neighbor approximation for RKKY interaction.

Up to this moment we treated the spin chains in a single-site approximation. This approximation is legitimate until $T \gg T_K \sim I$. To move further, we decouple the Euclidean action of the model (4), (5),

$$\mathcal{A} = \int_0^\beta \mathrm{d}\tau \left[\sum_j \left(\bar{c} \mathcal{G}_0^{-1} c + \bar{d} \mathcal{D}_0^{-1} d \right) - H_{int}^{cd} - H_{int}^{dd} \right] \tag{6}$$

by means of the Hubbard-Stratonovich scheme [15] in terms of the fields [9, 16]

$$\Delta_{j,j\pm1} \to \sum_\sigma \left(d_{j\sigma}^{\dagger} d_{j\pm1,\sigma} + \text{c.c.} \right), \qquad \phi_l \to \sum_{\mathbf{k}\sigma} \left(c_{l-1,\mathbf{k}\sigma}^{\dagger} (d_{j,\sigma} + d_{j+1,\sigma}) + \text{c.c.} \right).$$

Here $\mathcal{G}_0^{-1} = \partial_\tau - \epsilon(-i\nabla) + \mu$ and $\mathcal{D}_{loc}^{-1} = \partial_\tau - i\pi/(2\beta)$ are bare inverse single-particle Green's functions (GF) for conduction electrons and local spins, respectively, $\beta = 1/T$. The field ϕ describes the single-site Kondo screening and the field Δ stands for the spinon propagation along

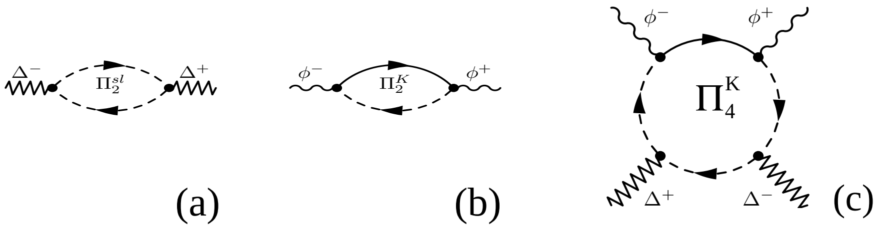

Fig. 2 – Loop expansion for non-local action (7). Solid and dashed lines in Π_2 and Π_4 stand for electron and spinon propagators, respectively.

the 1D spin chain with AFM coupling. The single occupancy constraint $d^\dagger_{j\uparrow}d_{j\uparrow}+d^\dagger_{j\downarrow}d_{j\downarrow}=1$ is preserved at each site in the chain by the semi-fermionic transformation [17]. These two fields resolve Doniach's dichotomy, because the long-range AFM order is absent in 1D.

We appeal to the uniform resonance valence bond (RVB) spin liquid state [16] and treat the spinon modes as fluctuations around the homogeneous solution in a nn- approximation, $\Delta_{j,j\pm1} \to = \Delta^u_{j,j\pm1} - \bar{\Delta}$ with $\bar{\Delta}^2(\beta)=\beta^{-1}\int_0^\beta \Delta(\tau)\Delta(-\tau)\mathrm{d}\tau$. For this sake, we add and subtract $\bar{\Delta}$ in the inverse GF. The non-local inverse spinon GF $\mathcal{D}^{-1} = \partial_\tau - \Delta_{j,j\pm1} - i\pi T/2$ has to be expanded in terms of $\Delta - \bar{\Delta}$. Now the two interacting components of bose-like modes in two-sublattice chain are spinons and Kondo clouds represented in effective action by $\Delta_{j,j+1}\Delta_{j+1,j}$ and $\phi_{l,l+m}\phi_{l+m,l}$ ($m = 0,1$), respectively. The charged ϕ-mode acquires dispersion due to the non-locality of $\tilde{J}_{lj}$, while the in-plane dispersion of conduction electrons in Kondo clouds is integrated out. The neutral spinon mode is dispersive by its origin. Then we come to an effective action with separated charge and spin sectors:

$$\mathcal{A}_{eff} = \sum_{jl,\omega_n}\left(\frac{|\phi_{l,l}(\omega_n)|^2}{\tilde{J}_{lj}} + \frac{|\Delta_{j,j+1}(\omega_n)|^2}{I}\right) + \\ +\mathrm{Tr}\log(\mathcal{G}_0^{-1}) + \mathrm{Tr}\log\left(\mathcal{D}^{-1}(\Delta_{j,j+1}) + \mathcal{G}_0\phi^*_{l,l}\phi_{l,l\pm1} + \text{c.c.}\right). \tag{7}$$

The last term in (7) may be represented as a loop expansion. The two first diagrams are shown in fig. 2. To calculate the diagrams, we use the non-local spinon GF $\mathcal{D}^0_{j,j+r}(\omega_n) = (\mathcal{D}^{-1}_{loc} - \bar{\Delta})^{-1}$ with cosine-like dispersion

$$\mathcal{D}^0_{j,j+r}(\omega_n) = \frac{\exp\left[-|r|\left[\ln\left(\frac{\bar{\Delta}}{\omega_n - \sqrt{\omega_n^2+\bar{\Delta}^2}}\right) - i\frac{\pi}{2}\right]\right]}{i\sqrt{\omega_n^2+\bar{\Delta}^2}}.$$

Here r numerates sites in the chain, $\omega_n = 2\pi T(n+1/4)$ on the imaginary axis [17]. $\mathcal{D}^0$ is characterized by a branch cut at $[-\bar{\Delta}, \bar{\Delta}]$. In the limit $\bar{\Delta} \ll \pi T$ it rapidly falls down with growing $|r|$ as $\mathcal{D}^0_{j,j+r}(\omega_n) \sim \bar{\Delta}^{|r|}/(i\omega_n)^{|r|+1}$. Thus the main contribution comes from $\mathcal{D}^0_{j,j}(\omega_n) = 1/i\sqrt{\omega_n^2+\bar{\Delta}^2}$ and $\mathcal{D}^0_{j,j\pm1}(\omega_n) = (\omega_n/\sqrt{\omega_n^2+\bar{\Delta}^2} - 1)/\bar{\Delta}$.

The polynomial effective action after the loop expansion acquires the form

$$\mathcal{A}_{eff} = \sum_{\langle jj'\rangle\omega_n}\left[\frac{|\Delta_{jj'}(\omega_n)|^2}{I} - \Pi_2^{sl}|\Delta_{jj'}(\omega_n) - \bar{\Delta}|^2\right] + \\ + \sum_{jj'l,\omega_n}\left(\frac{1}{\tilde{J}_{jl}} - \Pi_2^K + \Pi_4^K|\Delta_{jj'}(\omega_n)|^2\right)|\phi_{lj}(\omega_n)|^2 + \mathrm{Tr}\log(\mathcal{G}_0^{-1}) + \mathrm{Tr}\log[(\mathcal{D}^0)^{-1}] + \mathrm{O}(|\phi|^4). \tag{8}$$

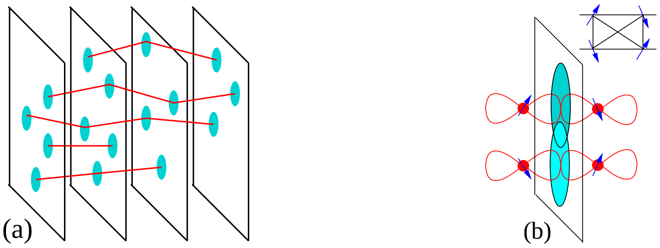

Fig. 3 – (a) Disordered anisotropic Kondo lattices. (b) Formation of spin ladder from interacting chains.

The polarization loops Π_2 and Π_4 are shown in fig. 2. The action (8) is gauge invariant in accordance with Elitzur theorem [16], and spin and charge modes are separated both in the 3D lattice and in the Fock space.

To estimate $\bar{\Delta}$, we refer to the properties of spin chains with AFM coupling [18]. The quasi–long-range order in these chains may be treated in terms of boson excitations in Luttinger liquid (LL) or fermion pairs in spin liquid. The spin susceptibility of a chain, $\langle \Delta^+ \Delta^- \rangle_{\omega=0} \sim \bar{\Delta}^2$, acquires Pauli form at $T^* \sim 8J^2/E_F$ [19], so we assume $\bar{\Delta} \sim T^*$ in our estimates. This means that even in the critical region of Doniach's diagram, $T_K \approx \tilde{J}^2/E_F$, the spins are "molten" into spin liquid at $T \sim T_K$, and there is no crossover to a strong Kondo coupling regime at low T.

Evaluation of Π_2, Π_4 in the limit $\pi T \gg \bar{\Delta}$ gives $\Pi_2^K \sim \rho_0 \ln(\bar{\Delta}/T)$ and $\Pi_4^K \sim \rho_0/\bar{\Delta}^2$. This leads to reduction of indirect exchange, $\tilde{I} = I[1+I/(\bar{\Delta}\ln(\bar{\Delta}/T_K))]^{-1}$. The main manifestation of weak Kondo screening in the LL limit at $T \to 0$ is the reduction of LL sound velocity, $\hbar v = \tilde{I} a_z$. As to the in-plane charge excitations, the formation of Kondo clouds is quenched at $T \gg T_K$, so instead of coherent Fermi liquid regime, $\langle \phi^+ \phi^- \rangle_{\omega \to 0}$ behaves as a relaxation mode $\sim [-i\omega/\Gamma + \alpha q^2 + \ln(\bar{\Delta}/T_K)]^{-1}$, where Γ, α are numerical constants.

These features of two-component electron/spin liquid manifest themselves in thermodynamics. The logarithmic corrections $\sim \ln^{-1}(T^*/T)$ are expected in low-T Pauli-like susceptibility of isotropic spin chains, whereas the log-corrections to the susceptibility of charged layers are quenched as $\ln(\bar{\Delta}/T_K)$. The overdamped relaxation mode should be seen as a quasielastic peak in χ_0. The 1D spinons contribute to the linear-T term in specific heat thus mimicking the heavy-fermion behavior, while the contribution of Kondo clouds is frozen at low T.

In real anisotropic crystals one may expect formation of distorted and dangling chains (fig. 3a) instead of an "ideal" lattice (fig. 1). Distortion means shift of two neighboring Kondo "shadows" in a stack. This effect may be modelled by a random overlap factor w_j in RKKY integrals, $I_j = w_j I$. The dangling bond effect means $w_j = 0$. Bond disorder may be treated in terms of random AFM chains [20]. According to this theory, the disorder results in transformation of singlet RVB liquid into a random-singlet RVB phase with arbitrarily long singlet bonds. More interesting effects associated with quantum criticality appear for $S = 3/2$ [21] where disorder-driven transition occurs between two different random singlet realizations. In the systems with integer spins of magnetic atoms the interplay between Haldane gap formation, effects of disorder and underscreened Kondo effect takes place. In chains with broken bonds the gaps arise due to the finite-length effect, so the short chain segments do not contribute to the low-T thermodynamics. With increasing impurity concentration, the Kondo clouds begin to overlap and two-leg ladders with diagonal bonds arise along with isolated chains (fig. 3b). Nozières' exhaustion is still not actual for these clusters. With further increase of the concentration of magnetic sites, Doniach's problem restores in its full glory. In case of FM coupling I, true long-range order emerges in spin chains, but Nozières' exhaustion is still quenched.

One may point out the class of layered conducting/magnetic hybrid molecular solids, where the possible candidates for the application of the above theory should be looked for. These crystals are formed by alternating metallic cationic layers and insulating magnetic anionic layers with various molecular groups as building blocks containing transition metal ions as carriers of localized spins [22]. Organic cations with magnetic ions in such systems form ordered stacks. The problem is in finding systems with metallic layers where the Kondo screening length $\hbar v_F/T_K$ is less than the distance between magnetic ions. It is worth noting, however, that the crystals containing dicyanamide radicals with Mn ions may form planar Kagome sublattice [22], thus being a promising object for the realization of the fractionalized Fermi liquid scenario proposed in [11].

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