Effect of Strong Intersite Correlations on the Mixing of Band and Localized States of the Periodic Anderson Model

V. V. Val'kov[a, b] and M. M. Korovushkin[a]

[a] *Kirensky Institute of Physics, Siberian Branch, Russian Academy of Sciences,*
Akademgorodok 50/38, Krasnoyarsk, 660036 Russia
[b] *Siberian Federal University, Krasnoyarsk, 660041 Russia*
e-mail: vvv@iph.krasn.ru
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It has been shown in the limit of the low density of band carriers that the inclusion of the strong intersite interaction between electrons within the framework of the periodic Anderson model results in two qualitatively new features in its energy structure. The first feature is associated with the appearance of an additional quasi-localized level, which significantly changes the structure of the electronic density of states. The second feature is the strong renormalization of the effective constant of hybridization with the bare localized level.

1. When studying the properties of strongly correlated systems within the framework of the periodic Anderson model [1], the Hubbard repulsion between two electrons that are located at one site and have opposite projections of spins is considered as a source of correlations, whereas the Coulomb interaction between two groups of electrons is disregarded. At the same time, the characteristic energy of the Coulomb interaction between electrons belonging to neighboring sites in many systems can insignificantly differ from the Hubbard energy. For such systems, the inclusion of intersite correlations in the calculation of the electronic structure can be important similarly to the inclusion of the single-ion interactions. The important role of intersite correlations was previously mentioned in [2], where the intermediate-valence phenomenon was analyzed, and in [3], where the effective Hamiltonian for copper oxides was constructed. It was pointed out recently that the intersite correlation effects leading to charge fluctuations can induce Cooper pairing [4].

In this paper, we report the results of investigations of the effect of intersite correlations on the electronic structure of the periodic Anderson model with the inclusion of the Coulomb interaction between two groups of electrons. It is assumed that f electrons are localized, whereas d electrons are collective and are described in the Wannier representation by hops over sites belonging to another sublattice.

To appropriately take into account the intersite correlations, the approximation beyond the Hartree–Fock approximation is used. The approach is based on the assumption that the collective electron density is low. In this case, the gas approximation is applicable and it is possible to trace the renormalizations of the energy structure of the periodic Anderson model that are caused by the intersite correlations.

2. To analyze the dynamics of the electrons in the crystal lattice of rare-earth compounds (see Fig. 1), we write the Hamiltonian of the periodic Anderson model including two strongly correlated subsystems of f and d electrons, as well as the Coulomb interaction between them. One-site correlations are taken into account by passing to the atomic representation. In this case, the Hamiltonian in the strong-correlation limit ($U \longrightarrow \infty$) can be represented in the form

$$\hat{H} = \sum_{m\sigma} E_0 Z_m^{\sigma\sigma} + \sum_{n\sigma} \varepsilon_d X_n^{\sigma\sigma}$$

$$+ V \sum_{mn\sigma} (Z_m^{\sigma 0} X_n^{0\sigma} + X_n^{\sigma 0} Z_m^{0\sigma}) \qquad (1)$$

$$+ t \sum_{nn'\sigma} X_n^{\sigma 0} X_{n'}^{0\sigma} + U_{fd} \sum_{mn} \hat{n}_m^f \hat{n}_n^d.$$

Here, the Hubbard operators Z_m^{pq} and X_n^{pq} [5] correspond to the transitions between the single-atomic states of two groups of electrons, the first and second terms describe the single-site energies of the electronic subsystems, the third term presents the hybridization of the f and d states, the fourth term describes the hops of electrons in the collective subsystem, and the fifth term corresponds to the intersite correlations of the two subsystems.

When $n_f \simeq 1$ and $n_d \ll 1$ per unit cell of the lattice under consideration, the interaction between collective electrons can be disregarded. Indeed, the characteristic energy of this interaction is quadratic in the small parameter n_d, $\sim U_{dd} n_d^2$, whereas the interaction between two subsystems is linear in n_d, $\sim U_{fd} n_f n_d \simeq U_{fd} n_d$. Thus, when the number of conduction electrons in the system is small, the problem of the energy spectrum contains the gas parameter $n_d \ll 1$. The use of this small parameter underlies the possibility of the correct description of the intersite correlations.

3. To calculate the energy spectrum of the system, we use the method of two-time temperature Green's functions [6] composed of the Hubbard operators [7, 8]. The exact equations of motion for the extended set of Green's functions have the form

$$(\omega - E_0)\langle\langle Z_m^{0\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega = \delta_{mm'}\langle Z_m^{00} + Z_m^{\sigma\sigma}\rangle$$

$$+ V\sum_n (\langle\langle (Z_m^{00} + Z_m^{\sigma\sigma})X_n^{0\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega \tag{2}$$

$$+ \langle\langle Z_m^{\bar\sigma\sigma} X_n^{0\bar\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega) + U_{fd}\sum_n \langle\langle Z_m^{0\sigma}\hat n_n^d|Z_{m'}^{\sigma 0}\rangle\rangle_\omega,$$

$$(\omega - E_0 - U_{fd})\langle\langle Z_m^{0\sigma}\hat n_n^d|Z_{m'}^{\sigma 0}\rangle\rangle_\omega$$

$$= \delta_{mm'}\langle (Z_m^{00} + Z_m^{\sigma\sigma})\hat n_n^d\rangle$$

$$+ V\sum_{\sigma_1} \langle\langle Z_m^{\sigma_1\sigma} X_n^{0\sigma_1}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega$$

$$+ V\sum_{m_1 \neq m}\sum_{\sigma_1} (\langle\langle Z_m^{0\sigma} X_n^{\sigma_1 0} Z_{m_1}^{0\sigma_1}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega \tag{3}$$

$$- \langle\langle Z_m^{0\sigma} Z_{m_1}^{\sigma_1 0} X_n^{0\sigma_1}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega)$$

$$+ t\sum_{n'}\sum_{\sigma_1} (\langle\langle Z_m^{0\sigma} X_n^{\sigma_1 0} X_{n'}^{0\sigma_1}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega$$

$$- \langle\langle Z_m^{0\sigma} X_{n'}^{\sigma_1 0} X_n^{0\sigma_1}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega),$$

$$(\omega - \varepsilon_d)\langle\langle X_n^{0\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega$$

$$= V\sum_m (\langle\langle (X_n^{00} + X_n^{\sigma\sigma})Z_m^{0\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega$$

$$+ \langle\langle X_n^{\bar\sigma\sigma} Z_m^{0\bar\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega)$$

$$+ t\sum_{n'} (\langle\langle (X_n^{00} + X_n^{\sigma\sigma})X_{n'}^{0\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega \tag{4}$$
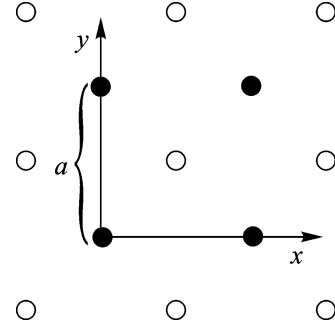

Fig. 1. Crystal lattice of a rare-earth intermetallide.

$$+ \langle\langle X_n^{\bar\sigma\sigma} X_{n'}^{0\bar\sigma}|Z_{m'}^{\sigma 0}\rangle\rangle_\omega)$$

$$+ U_{fd}\sum_m \langle\langle X_n^{0\sigma}\hat n_m^f|Z_{m'}^{\sigma 0}\rangle\rangle_\omega.$$

This system of equations contains higher Green's functions in which the left operator is the product of different-site operators. The energy factors upon which these functions are multiplied are significantly different when $U_{fd} \gg V, t$. Furthermore, when Hubbard's idea [9] proposed for taking into account one-site correlations is generalized to the intersite correlations, it is seen that the normal uncoupling of the Hartree–Fock approximation type can be performed in the Green's functions that are not multiplied by the large energy parameter U_{fd}. At the same time, the intersite Green's functions appearing in the equations with a large energy factor should be taken into account exactly and the corresponding equations of motion should be written for them. The number of such new equations in our case decreases significantly due to the use of the gas approximation. In this approximation, all intersite Green's functions describing more than one collective electron near a rare-earth ion are disregarded.

In the linear approximation, the spin-flip processes can be ignored and only spatially homogeneous solutions can be used. The simplest approximation also allows one to disregard the kinetic correlation functions $\langle Z_m^{\sigma 0} X_n^{0\sigma}\rangle$ and $\langle X_n^{\sigma 0} X_n^{0\sigma}\rangle$. In this case, the closed system of equations for Green's functions in the quasi-momentum representation can be simply reduced to the form

$$(\omega - E_0)\langle\langle Z_{\mathbf{k}\sigma}|Z_{\mathbf{k}\sigma}^\dagger\rangle\rangle$$

$$= \tilde N_f(\omega) + V_{\mathbf{k}} A(\omega)\langle\langle X_{\mathbf{k}\sigma}|Z_{\mathbf{k}\sigma}^\dagger\rangle\rangle, \tag{5}$$

$$(\omega - \varepsilon_{\mathbf{k}})\langle\langle X_{\mathbf{k}\sigma}|Z_{\mathbf{k}\sigma}^\dagger\rangle\rangle = V_{\mathbf{k}} N_d \langle\langle Z_{\mathbf{k}\sigma}|Z_{\mathbf{k}\sigma}^\dagger\rangle\rangle.$$

$$\tilde N_f(\omega) = N_f\left(1 + \frac{4U_{fd}n_d}{(\omega - E_0 - U_{fd})}\right), \quad N_f = 1 - \frac{n_f}{2},$$
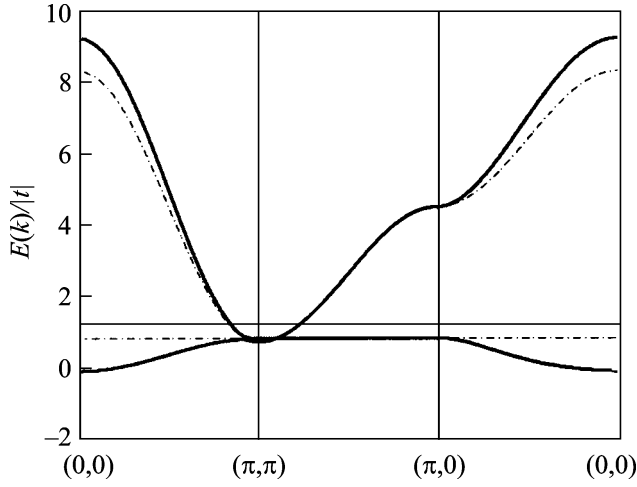
Fig. 2. Band structure of the periodic Anderson model in the Hartree–Fock approximation ($n_f = 0.97$, $n_d = 0.10$, and $\mu = 1.2$).

$$A(\omega) = 1 - \frac{n_f}{2} + \frac{n_f}{2}\frac{U_{fd}}{(\omega - E_0 - U_{fd})}, \quad N_d = 1 - \frac{n_d}{2},$$

$$\varepsilon_{\mathbf{k}} = \tilde{\varepsilon}_d + t_{\mathbf{k}}N_d, \quad V_{\mathbf{k}} = 4V\cos\left(\frac{k_x\mathbf{a}}{2}\right)\cos\left(\frac{k_y\mathbf{a}}{2}\right),$$

$$\tilde{\varepsilon}_d = \varepsilon_d + 4U_{fd}, \quad t_{\mathbf{k}} = 2t(\cos(k_x\mathbf{a}) + \cos(k_y\mathbf{a})).$$

This system of equations includes a number of features. First, the coefficients of the desired Green's functions are functions of the frequency. This induces renormalizations depending both on the total number of electrons in the system and on the energy range where the Fermi excitations under consideration are realized. In particular, the hybridization of two electronic subsystems near the lower pole $E \simeq E_0$ at $U_{fd} \neq 0$ is suppressed when $n_f \longrightarrow 1$. This is mathematically expressed in the form of the renormalization of the hybridized interaction as $V \longrightarrow \tilde{V} = V(1 - n_f)$. In other words, the lower energy band corresponding to the localized electronic states becomes dispersionless at $n_f \longrightarrow 1$ and this behavior can give rise to a significant increase in the effective mass of the fermions. Second, the collective electron band is shifted by $4U_{fd}$. The physical cause of this shift is obvious: one band electron appearing at the site of the lattice interacts with four electrons localized at neighboring sites.

According to system of equations (5), the spectrum of Fermi excitations is determined by the solution of the cubic equation

$$E^3 - mE^2 + pE + q = 0, \tag{6}$$

where

$$m = \varepsilon_{\mathbf{k}} + 2E_0 + U_{fd},$$

$$p = \varepsilon_{\mathbf{k}}(2E_0 + U_{fd}) + E_0(E_0 + U_{fd}) - N_fN_dV_{\mathbf{k}}^2, \tag{7}$$

$$q = (E_0 + U_{fd})(N_fN_dV_{\mathbf{k}}^2 - E_0\varepsilon_{\mathbf{k}}) - U_{fd}N_d\left(\frac{n_f}{2}\right)V_{\mathbf{k}}^2.$$

Equation (6) has three roots

$$E_{1\mathbf{k}} = -\frac{s}{2} + t + u + i\frac{\sqrt{3}}{2}(s + 2t),$$

$$E_{2\mathbf{k}} = -\frac{s}{2} + t + u - i\frac{\sqrt{3}}{2}(s + 2t), \tag{8}$$

$$E_{3\mathbf{k}} = s - 2t + u,$$

where

$$s = \frac{1}{6}(-36mp - 108q + 8m^3$$

$$+ 12\sqrt{12p^3 - 3m^2p^2 + 54mpq + 81q^2 - 12qm^3})^{1/3}, \tag{9}$$

$$t = \frac{3p - m^2}{18s}, \quad u = \frac{m}{3}.$$

With the use of the spectral theorem [10], it is easy to write the system of self-consistency equations to obtain the dependence of the energy structure on the number of electrons in the system:

$$\frac{n_f}{2} = \frac{1}{N}\sum_{\mathbf{k}}\sum_i A_i(E_{i\mathbf{k}})f(E_{i\mathbf{k}}),$$

$$\frac{n_d}{2} = \frac{1}{N}\sum_{\mathbf{k}}\sum_i B_i(E_{i\mathbf{k}})f(E_{i\mathbf{k}}), \tag{10}$$

$$n_f + n_d = n.$$

Here,

$$A_i(E_{i\mathbf{k}}) = \sum_{i=1}^{3}\frac{\tilde{N}_f(E_{i\mathbf{k}})(E_{i\mathbf{k}} - \varepsilon_{\mathbf{k}})(E_{i\mathbf{k}} - E_0 - U_{fd})}{\prod_{j\neq i}(E_{i\mathbf{k}} - E_{j\mathbf{k}})},$$

$$B_i(E_{i\mathbf{k}}) = \sum_{i=1}^{3}\frac{N_d(E_{i\mathbf{k}} - E_0)(E_{i\mathbf{k}} - E_0 - U_{fd})}{\prod_{j\neq i}(E_{i\mathbf{k}} - E_{j\mathbf{k}})}, \tag{11}$$

$i = 1, 2, 3$, are the spectral intensities, $f(x) = \left(\exp\left(\frac{x - \mu}{T}\right) + 1\right)^{-1}$ is the Fermi–Dirac function, and n is the total number of electrons.

4. For comparison, Fig. 2 shows the well-known band structure of the model under consideration in the
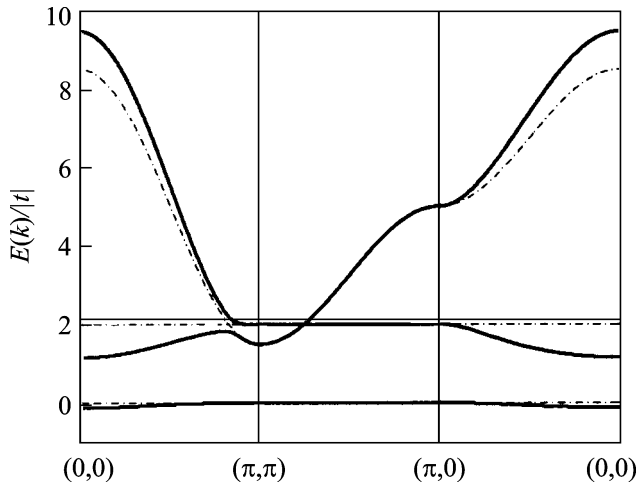
Fig. 3. Band structure of the periodic Anderson model with the inclusion of intersite correlations in the gas approximation ($n_f = 0.95$, $n_d = 0.24$, and $\mu = 2.12$).
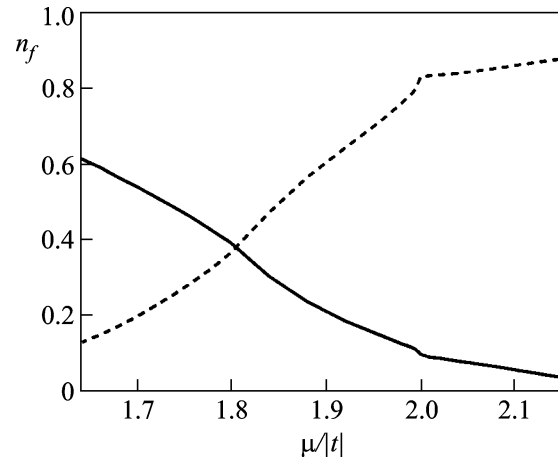

Fig. 4. Chemical-potential dependence of localized-electron densities in the (solid line) lower and (dashed line) upper f subbands.

Hartree–Fock approximation for the following parameters:

$$t = 1 \text{ eV}, \quad E_0 = 0,$$

$$\frac{\tilde{\varepsilon}_f}{|t|} = 5, \quad \frac{V}{|t|} = 1, \quad \frac{U_{fd}}{|t|} = 2. \tag{12}$$

The thick solid lines present the resulting spectrum of Fermi excitations and the dashed lines show the bare spectrum. The thin solid horizontal straight line marks the chemical potential of the system, μ. It is seen that the energy structure of the system consists of two bands: the lower band is formed predominantly by localized f states and the upper band corresponds to collective d states. Figure 3 shows the energy spectrum of the model calculated with the inclusion of the intersite correlations in the gas approximation for the same parameters given in (12). A comparison shows that the qualitative difference between the two spectra shown in Figs. 2 and 3 is that the formation of the latter spectrum involves not one, but two split localized levels spaced by U_{fd}. The lower subband corresponds to localized f electrons near which band electrons are absent. The upper subband corresponds to states with a higher excitation energy $E_0 + U_{fd}$, which is due to the appearance of a band electron near a localized electron. Note that the mechanism of the splitting of the localized level into two levels is qualitatively similar to the mechanism of the appearance of two Hubbard subbands in the presence of a strong interatomic repulsion. Indeed, the lower band in the Hubbard model corresponds to the motion of an electron over sites at which other electrons are absent. The upper band appears due to hops over the sites at which electrons with opposite spin projections are located. For this reason, the energy of such a band should include the shift by the Coulomb repulsion energy U. Similarly, when collective states are free, the

appearance of an electron in a localized state corresponds to an increase in the energy by ε_f. If one collective electron is located near an f level, the energy $\varepsilon_f + U_{fd}$ is required to occupy this f level. Thus, the mechanism of the appearance of the two-level structure of localized states is a simple generalization of the mechanism of the appearance of two Hubbard subbands with the inclusion of one-site correlations to the case of the inclusion of the intersite correlations.

This conclusion is confirmed in Fig. 4, where the populations of the lower and upper levels are shown as functions of the chemical potential. We recall that, owing to the electron-density dependence of the spectral intensities of the Hubbard subband in the Hubbard model, doping is accompanied by the redistribution of the spectral weight between these subbands. In our case, a similar behavior is observed: as the number of electrons in the system increases, the localized electrons are redistributed between the lower and upper f subbands. As seen in Fig. 4, when μ increases, the number of f electrons with the characteristic energy E_0 decreases, whereas the number of f electrons with the energy $E_0 + U_{fd}$ increases. At $\mu = 2.2$, the number of f electrons with the characteristic energy E_0 vanishes and almost all localized electrons are in the upper f subband. In addition, a small fraction of f electrons appears in the conduction band due to hybridizing effects. The reported results indicate that the energy spectrum of the Fermi excitations in the presence of strong intersite correlations is formed due to hybridizing processes with the upper localized level. This circumstance qualitatively changes the energy structure of the periodic Anderson model. It is substantial that the effective hybridization with the lower of the split localized states is strongly suppressed due to the renormalization mentioned above. This gives rise to the appearance of the
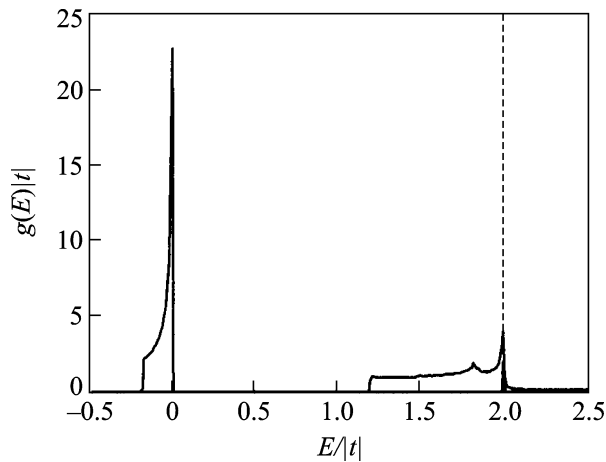
Fig. 5. Electronic density of states ($n_f = 0.92$ and $n_d = 0.23$). The vertical dashed straight line shows the chemical potential $\mu = 2$.

Fig. 6. Chemical potential versus the total electron number.

peak in the electronic density of states near the lower level (see Fig. 5). The density of states near the second level also increases significantly (this increase is smaller than the increase in the density of states near the lower level, but is noticeably larger than the increase in the density of states in the collective band). We emphasize that the increase in the density of states near the chemical potential is obtained without the use of the slave-boson representation.

The results of the numerical analysis of self-consistency equations (10), which, in particular, present the relation between the total number of electrons in the system and the filling factor of the energy levels, are presented in Fig. 6, where the chemical potential is shown as a function of the total number of fermions. It is seen that the chemical potential of the system where the number of electrons per unit cell is slightly smaller than one is in the upper f subband. An increase in the number of electrons leads to an increase in the chemical potential and to an intensive occupation of the upper subband of the quasi-localized states. A kink is seen in the density dependence of the chemical potential at $\mu = 2$. This kink is attributed to the beginning of the filling of the collective band of p states. The difference between the slopes in these dependences presents the difference between the densities of states in the collective band and in the upper collective subband.

To conclude, we note that the method proposed for taking into account strong intersite Coulomb correlations is a natural generalization of an approach actively used in the theory of strongly correlated systems with one-site correlations. Its consistent application shows that strong intersite correlations not only induce 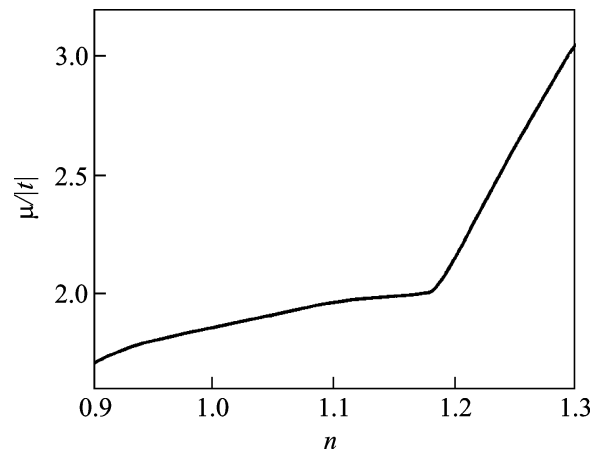quantitative changes in the electronic structure of the periodic Anderson model, but also qualitatively modify, for example, the density of states. This means that the inclusion of intersite correlations is substantial for the interpretation of the thermodynamic properties of rare-earth intermetallides described by the periodic Anderson model.

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