Damped Ruderman-Kittel-Kasuya-Yosida interaction among local magnetic moments in the impurity-band regime of doped semiconductors

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We consider a highly doped semiconductor with a metallic impurity band at $T = 0$ K. A small number of localized spins is assumed to exist as well, with a site concentration much smaller than that of the dopant impurities. The carriers in the impurity band are described in an independent-particle tight-binding approximation. The effective interaction among the localized spins is expressed in terms of the spin susceptibility of the impurity band, which is obtained in turn by Green-function perturbative techniques within an approximation due to Matsubara and Toyozawa, asymptotically valid at high concentration. The range function of the effective interaction shows a damped oscillatory Ruderman-Kittel-Kasuya-Yosida form, with a wavelength of the order of the average separation of dopant impurities. Exponential decay is a consequence of the disorder of the one-electron impurity potential. The results are relevant in connection with the low-temperature magnetic properties of heavily doped semiconductors and also for magnetic multilayers with such systems as separators.

I. INTRODUCTION

As is well known, Si and Ge doped with several atomic species of valence different from four, exhibit shallow impurity states. As the impurity concentration increases and reaches a critical n_c, an insulator-metal transition (MIT) is observed, and on the metallic side, for $n > n_c$, the electronic properties at low T are dominated by carriers (electrons or holes, depending on whether the doping is n or p type, respectively), which form an *impurity band* of states. As n increases further, a second kind of phase transition is observed in which the Fermi level in the impurity band, which increases with n, reaches for $n = n_{cb}$ the bottom of the corresponding empty band (conduction band for electrons, valence band for holes) of the host semiconductor, and the electronic properties for $n > n_{cb}$, are dominated by the states of the bottom of that band, in the presence of a random potential produced by the impurities.[1]

Recent experiments have dealt with the change of the magnetic properties at low T as a function of n, for n near n_c.[2,3] The conclusion seems to be that local magnetic moments exist at $n < n_c$, and persist above n_c, into the disordered metallic phase, where they seem to dominate the low T thermodynamic properties and the spin dynamics.

The idea that in a few percent of the impurities there are local conditions (local electron concentration and effective Coulomb repulsion), which allow for nonvanishing local moments, seems to be in agreement with experimental results on Si:P. The ESR studies of boron compensated Si:P near the MIT support a model in which both local moments and itinerant (impurity band) electrons are present. Also, susceptibility measurements[3] show a divergent susceptibility as $T \to 0$, indicating the presence of localized spins.

The existence of local moments in the metallic phase was already suggested by early measurements[4] and a first theoretical explanation was advanced by Toyozawa[5] within a Hartree-Fock formulation. A recent theoretical simulation of the disordered impurity band in the Hartree-Fock approximation[6] indicates that the percentage of dopant sites with nonvanishing local moments would vary from 1% at $p = 3$ to 16% at $p = 1$, in terms of a dimensionless dopant concentration defined as

$$p = 32\pi n(a^*)^3,$$

where n is the volume dopant concentration and a^* is the effective Bohr radius of the shallow impurity wave functions. In these units, the MIT would occur at $p_c \cong 1.6$, according to Mott's criterium.

The experiments indicate that an effective exchange interaction exists among the local moments. This interaction was estimated in numerical simulations,[3] for the insulating phase, as due to direct exchange among electrons on neighbor impurity sites. On the metallic side of the MIT, the expected concentration of local spins should be very small, thereby ruling out the process of direct exchange among them. In this case, we could consider as an alternative the possibility of indirect exchange interaction among those local moments that still survive in this phase.

This consideration leads us to discuss as well the possibility that magnetic layer systems separated by such a heavily doped semiconducting layer might interact through the same effective indirect exchange. In the case

of magnetic multilayers, the local spins are those belonging to the magnetic layers, so that any complete description of this problem should include some model for the magnetic-metal/semiconductor interface.

In the present work, we consider that the semiconductor under study is doped with only one impurity species, that the impurities are distributed randomly, and that only a few percent of the sites exhibit local moments. We study the indirect exchange interaction among these localized spins via itinerant electrons of the impurity band, at zero temperature, using a tight-binding Hamiltonian within a hydrogenlike $1s$ basis. Our results show that this interaction has a damped Ruderman-Kittel-Kasuya-Yosida (RKKY) range function.

II. MODEL AND METHOD OF CALCULATION

A. The impurity-band problem

As a first approximation, we apply to this problem the results of a classical paper by Matsubara and Toyozawa (MT) (Ref. 7) on disordered electronic systems. Their model assumes that the impurities are distributed completely at random in the semiconductor host lattice. At each impurity site, according to the effective mass approximation, one obtains the usual shallow impurity states. We shall adopt the simplest possible approximation for these localized states, and consider them $1s$ hydrogenic-type states with an effective Bohr radius a^*, which can be typically from 10 to 50 lattice constants. Using this basis, MT consider a tight-binding Hamiltonian,

$$\mathcal{H} = \sum_m \sum_{\neq n} \sum_\sigma V_{mn} a^\dagger_{m\sigma} a_{n\sigma}, \tag{1}$$

where V_{mn} is the hopping integral from site n to site m, and $a^\dagger_{n\sigma}$ and $a_{n\sigma}$ are, respectively, the creation and annihilation operators of an electron with spin σ at the corresponding site.

The Hamiltonian can be diagonalized by an orthogonal transformation,

$$a_{m\sigma} = \sum_\mu c^\sigma_{m\mu} A_{\mu\sigma} \tag{2}$$

where $A_{\mu\sigma}$ is the annihilation operator for the μth eigenstate of an electron with σ spin in the presence of all the impurities, in a given fixed configuration.

In the tight-binding approach, the spin operators at the position of the impurity R_m are

$$\sigma^\pm(\vec{R}_m) = a^\dagger_{m\pm} a_{m\mp}. \tag{3}$$

Let us imagine now, following the ideas summarized in the Introduction, that a few percent of the impurity sites have a nonzero local magnetic moment in the metallic phase, in which the impurity band is occupied by electrons with energies up to the Fermi level.

B. Generalized RKKY interaction

Let us consider in what follows an n-type sample, so that the impurities are donors and the majority carriers are electrons, and let us neglect the effect of the compensating impurities, if present. We assume that at some sites $\{\vec{R}_\alpha\}$ local moments exist, which we shall represent by spin operators $\{\vec{S}_\alpha\}$.

As in the case of noble metals with paramagnetic impurities, let us assume a local Friedel exchange interaction,[8]

$$V(\vec{R}_n) = J_0 \vec{S}_{\vec{R}} \cdot \vec{\sigma}_n \, \delta_{\vec{R},\vec{R}_n}, \tag{4}$$

between a conduction electron at $\vec{R}_n$, with spin $\vec{\sigma}_n$ and a local spin at site $\vec{R}$, $\vec{S}_R$. Equation (4) can be written as

$$V(\vec{R}_n) = -\vec{m}_n \cdot \vec{B}(\vec{R}_n), \tag{5}$$

where the electronic magnetic moment $\vec{m}_n = \gamma\vec{\sigma}(\vec{R}_n)$ and the external field, due to the local spin $S(\vec{R})$ at $\vec{R}_n$ is

$$\vec{B}(\vec{R}_n) = -\frac{J_0}{\gamma} \vec{S}_{\vec{R}} \, \delta_{\vec{R},\vec{R}_n}. \tag{6}$$

In (6) we write a Kronecker δ, since the set of sites is discrete.

In the static limit of the linear response theory, we can express the induced static magnetic moment at the impurity site R_α as

$$\vec{m}(\vec{R}_\alpha) = \gamma^2 \sum_\beta \int_0^\infty \chi(\vec{R}_\alpha, \tau; \vec{R}_\beta, 0) \, \vec{B}(\vec{R}_\beta) \, d\tau, \tag{7}$$

where the sum extends over all magnetic sites, which act as field sources.

We chose convenient units for χ in (7) by introducing the factor γ^2. The fluctuation-dissipation theorem yields for the response function $\gamma^2\chi$ the expression:

$$\gamma^2 \chi^{ab}(\vec{R}_\alpha, \tau; \vec{R}_\beta, 0) = -i\theta(\tau)\langle[m_a(\vec{R}_\gamma, \tau), m_b(\vec{R}_\beta, 0)]\rangle, \tag{8}$$

where (a, b) stand for the cartesian or circular space components, and $\langle A\rangle$ is the thermodynamic canonical average.

If another local static spin is at site $\vec{R}'$, the potential energy of interaction with the induced electronic spin at site $\vec{R}_l$ is

$$\begin{aligned} V(\vec{R}_l, \vec{R}') &= \frac{J_0}{\gamma} \vec{S}_{\vec{R}'} \cdot \vec{m}(\vec{R}_l) \, \delta_{\vec{R}',\vec{R}_l} \\ &= J_0 \gamma \vec{S}_{\vec{R}'} \cdot \sum_\beta \int_0^\infty \chi(\vec{R}_l, \tau; \vec{R}_\beta, 0) \left(\frac{-J_0}{\gamma} \vec{S}_{\vec{R}}\right) d\tau \, \delta_{\vec{R},\vec{R}_\beta} \delta_{\vec{R}',\vec{R}_l} \\ &= -J_0^2 \, \delta_{\vec{R}',\vec{R}_l} \vec{S}_{\vec{R}'} \cdot \sum_\beta \vec{S}_{\vec{R}_\beta} \int_0^\infty \chi(\vec{R}_l, \tau; \vec{R}_\beta, 0) d\tau), \end{aligned} \tag{9}$$

where we assume that χ is isotropic, as discussed below. Summing over all magnetic sites, we get the total interaction energy among local magnetic moments as an effective spin-spin interaction thereof. In the static limit this interaction is

$$W = \frac{1}{2}\sum_{\alpha\neq\beta} J_{\rm eff}(\vec{R}_\alpha,\vec{R}_\beta)\,\vec{S}_\alpha\cdot\vec{S}_\beta, \tag{10}$$

with

$$J_{\rm eff}(\vec{R}_\alpha,\vec{R}_\beta) = -J_0^2\int_0^\infty \chi(\vec{R}_\alpha,\tau;\vec{R}_\beta,0)d\tau \tag{11}$$

and by appeal to (8), we have

$$\chi^{ab}(\vec{R}_\alpha,\tau\,;\vec{R}_\beta,0) = -i\theta(\tau)\langle[\sigma_a(\vec{R}_\alpha,\tau),\sigma_b(\vec{R}_\beta,0)]\rangle. \tag{12}$$

Since χ is assumed isotropic, which it will necessarily be after averaging over impurity configurations, we calculate only the transverse component,

$$\chi^{-+}(\vec{R}_n,\tau;\vec{R}_m,0) = -i\theta(\tau)\langle[\sigma^-(\vec{R}_n,\tau),\sigma^+(\vec{R}_m,0)]\rangle, \tag{13}$$

where

$$\begin{aligned}\sigma^+(\vec{R}_n,\tau)&=\sum_{\mu,\nu}\Omega^n_{\nu,\mu}A^\dagger_{\nu+}(\tau)A_{\mu-}(\tau),\\ \sigma^-(\vec{R}_n,\tau)&=\sum_{\mu,\nu}\Omega^n_{\mu,\nu}A^\dagger_{\mu-}(\tau)A_{\nu+}(\tau),\\ \Omega^m_{\mu\nu}&=c^*_{m\mu}c_{m\nu},\end{aligned} \tag{14}$$

[see Eqs. (2) and (3)].

Upon substitution of (14) into (13), we are led to the calculation of commutators of productors of operators $A_{\mu\sigma}$, which are easily performed since they obey fermion anticommutation relations:

$$\{A^\dagger_{\mu\sigma},A_{\nu\sigma'}\}=\delta_{\mu\nu}\delta_{\sigma\sigma'}. \tag{15}$$

The result is

$$\chi^{-+}_{nl}(\tau) = -i\theta(\tau)\sum_{\mu,\nu}\Omega^n_{\mu\nu}\Omega^l_{\nu\mu}\,[f_\mu(1-f_\nu) - f_\nu(1-f_\mu)]\,e^{i\frac{\tau}{\hbar}(E_\mu-E_\nu)}, \tag{16}$$

with the notation

$$f_\mu = \frac{1}{e^{\beta(E_\mu-E_F)}+1}. \tag{17}$$

At this point, we follow MT and introduce the one-particle Green's functions,

$$\begin{aligned}G^\pm_{nm\sigma}(\tau) &= \mp i\langle n\mid e^{-i(H\mp i\varepsilon)\tau}\mid m\rangle\theta(\pm\tau)\\ &= \mp i\sum_\mu c^*_{n\mu}c_{m\mu}e^{-i(E_\mu\mp i\varepsilon)\tau}\,\theta(\pm\tau),\end{aligned} \tag{18}$$

which are spin independent in the present approximation. + (−) stand for retarded (advanced) assymptotic conditions. G^+ (G^-) is analytic in the upper (lower) half plane of the energy. In fact, the Fourier transform of (18) is

$$G^\pm_{nm}(E) = \sum_\mu \frac{c^*_{n\mu}c_{m\mu}}{E-E_\mu\pm i\varepsilon}. \tag{19}$$

Matsubara and Toyozawa also define

$$\begin{aligned}G_{nm}(E) &= \frac{1}{2\pi i}[G^+_{nm}(E)-G^-_{nm}(E)]\\ &= -\sum_\mu \delta(E-E_\mu)c^*_{n\mu}c_{m\mu},\end{aligned} \tag{20}$$

which for $n=m$ gives minus the local density of states.

We now integrate Eq. (16) with respect to τ, to obtain the Fourier transform for $\omega=0$ (static limit), and we also introduce a double integration on energy:

$$\chi^{-+}_{nl}(0) = \sum_{\mu,\nu}\int dE\int dE'\frac{(f(E)-f(E')}{E-E'+i\varepsilon}\,\delta(E-E_\mu)\,\delta(E'-E_\nu)\,\Omega^n_{\mu\nu}\Omega^l_{\nu\mu}, \tag{21}$$

which can be rewritten in terms of G_{ln}, G_{nl} as

$$\chi{-+}_{nl}(0) = \int dE\int dE'\frac{f(E)-f(E')}{E-E'+i\varepsilon}\times G_{nl}(E)\,G_{ln}(E'). \tag{22}$$

Upon performing one energy integration, we get

$$\chi^{-+}_{nl}(0) = -\frac{1}{\pi}\int_{-\infty}^{\infty} dE f(E)\,{\rm Im}[(G^+_{ln}(E))^2]. \tag{23}$$

This function of $\vec{R}_n$ and $\vec{R}_l$ is the generalization of the RKKY range function $\Phi(\mid\vec{R}-\vec{R}_l\mid)$ (Ref. 8) to an arbitrary spatial distribution of atoms, within the tight-binding, independent-particle approximation. One recovers the well known expression if one assumes a perfectly ordered lattice of sites, and substitutes the one-electron propagators for a crystal into (23).

In the rest of this section, we summarize the procedure for finding the configuration average of (23), following the MT formalism. Let us stress, however, that (23) is the correct expression for χ in the present approximation, for a fixed configuration, and it can be used to obtain the probability distribution of χ, and accordingly of the range function, without recourse to the further drastic approximations involved in the MT procedure for obtaining averages over configurations. In what follows, a bar over a quantity will denote its configurational average.

Matsubara and Toyozawa developed a graphical method to treat the spatial disorder in the semiconductor and they obtained the diagonal (G_{nn}) and nondiagonal (G_{nm}) Green-function matrix elements, in terms of the self-consistent solutions ($\xi^\pm,\zeta^\pm$) of the equations:

$$(1-w_\pm)+\frac{w_\pm}{\xi^\pm} = \frac{8}{\pi}\int_{-\infty}^{\infty}\frac{t^2\,dt}{(t^2+1)^3+(p\xi^\pm/w_\pm)}, \tag{24}$$

$$\zeta^\pm(r,w) = -\frac{(\xi^\pm)^2}{w_\pm}\frac{8}{\pi ir}\int_{-\infty}^{\infty}\frac{t\,e^{itr}\,dt}{(t^2+1)^3+(p\xi^\pm/w_\pm)}, \tag{25}$$

with the dimensionless parameters,

$$w_{\pm} = \frac{E \pm i\varepsilon}{V_0},$$
$$r = \frac{R}{a^*}, \tag{26}$$

where V_0 is twice the ionization energy of the $1s$-state, $R = \mid \vec{R}_n - \vec{R}_m \mid$ and p and a^* have been defined in the introduction.

The quantities ξ and ζ are defined as

$$\xi^{\pm}(E) = E\,\overline{G^{\pm}_{nn}(E)},$$
$$\zeta^{\pm}(E) = E\,\overline{G^{\pm}_{nm}(E)}, \tag{27}$$

and they are dimensionless.

We further make the approximation,

$$\overline{G^{+}_{mn}(E)\,G^{+}_{nm}(E)} \approx \overline{G^{+}_{mn}(E)}\;\overline{G^{+}_{nm}(E)} \tag{28}$$

(which neglects the interference of electron and hole propagating in the random potential), so we can rewrite the average of (23) as

$$\chi^{-+}(\omega; nm)|_{\omega=0} = -\frac{1}{\pi V_0}\int_{-\infty}^{w_F} \frac{\mathrm{Im}((\zeta^{+})^2)}{w^2}\,dw. \tag{29}$$

The MT approximation is adequate for $p \gg 1$, as regards the one-particle propagators. The further factorization (28) is only made for simplicity. It has been used with success by MT to calculate the conductivity.

III. RESULTS AND CONCLUSIONS

We have one varying parameter in this problem, namely, the dopant concentration n, which leads to a characteristic length $d = n^{-1/3}$. It is, therefore, interesting to use d as unit length.

We fit the numerical results with the expression

$$\chi = \alpha \frac{e^{R/l}}{R^3}\cos(R/\lambda). \tag{30}$$

The results of the self-consistent calculation, for the static electronic magnetic susceptibility χ^{-+}, as a function of the distance between localized sites, in units of d, are shown in Fig. 1 for $p = 1$ and in Fig. 2 for $p = 7$. We also indicate the values of R/a^* on the horizontal axis in both figures. At all concentrations, χ oscillates with a well defined wavelength λ, which scales approximately as

$$\lambda = Cp^{-\frac{1}{3}}, \tag{31}$$

as shown in Fig. 3 in a log-log plot. The straight line is the best linear fit to the numerical results.

Fitting the amplitude, on the other hand, leads to minimum square fit results for $\alpha(p)$ and $l(p)/d$ shown in Figs. 4 and 5, respectively. The length l can be inter-

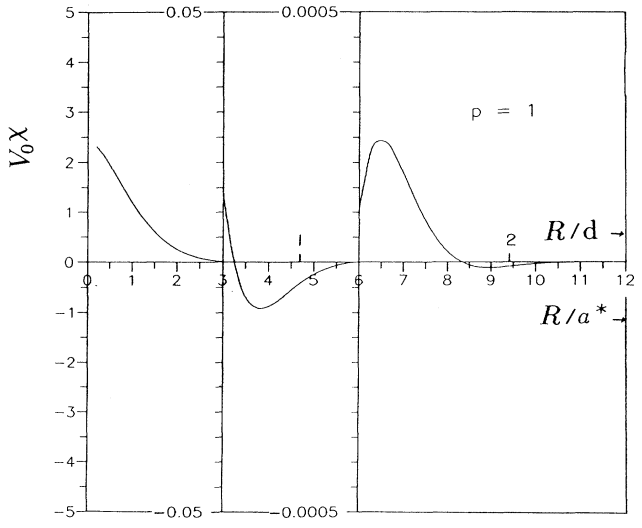

FIG. 1. Dimensionless susceptibility $V_0\chi$ as a function of R/d for $p = 1$. We display also the values of R/a^* for reference on the horizontal scale.

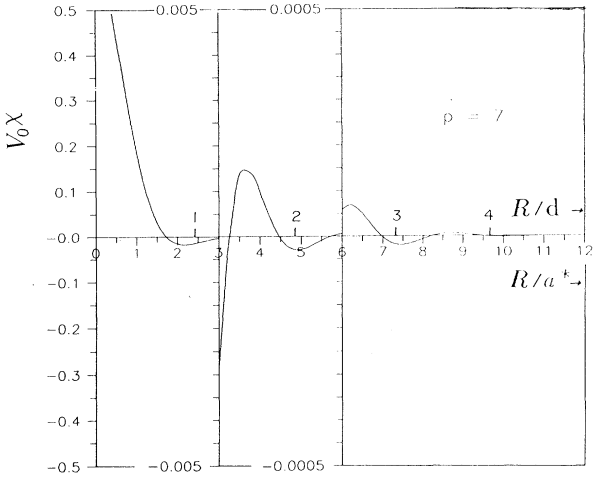

FIG. 2. Same as Fig. 1, for $p = 7$.

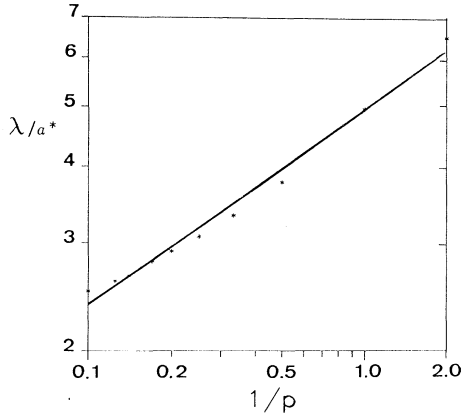

FIG. 3. Wavelength λ of the susceptibility as a function of $1/p$ in units of the effective Bohr radius a^*, in a log-log plot.

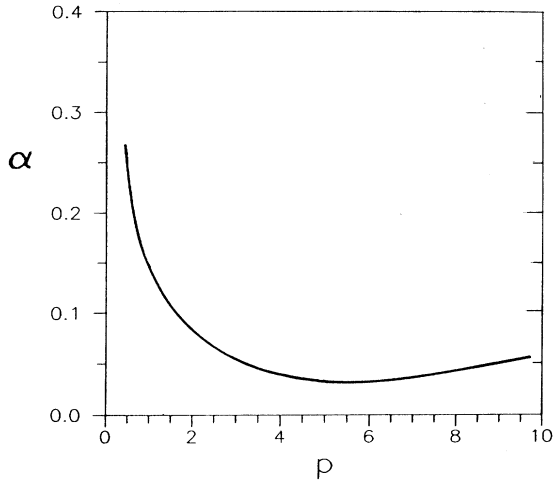

FIG. 4. Plot of coefficient α of Eq. (30) vs p.

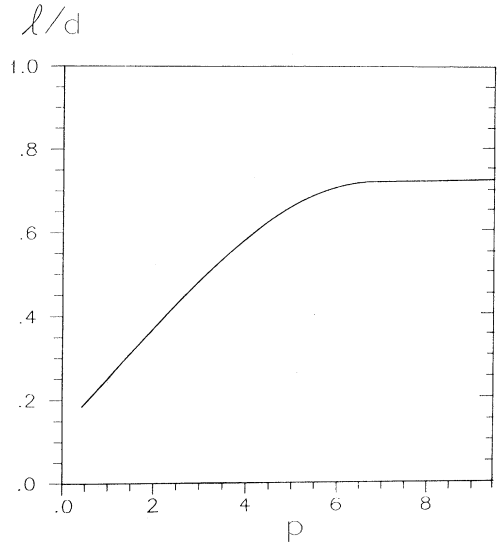

FIG. 5. Parameter l of Eq. (30) vs p, in the units of d.

preted as an average localization length of electrons and holes in the disordered potential.

We see that both l and α saturate approximately for $p > 4$. Let us remind you that the MIT for Si:P occurs experimentally at $p = 0.8$, which corresponds to $n_c = 1.95 \times 10^{18}$ cm^{-3}, while at the second transition mentioned at the Introduction $n_{cb} = 1.70 \times 10^{19}$ cm^{-3} and $p = 7$. The oscillating behavior of χ is qualitatively consistent with early results for the off diagonal $\overline{G_{nl}(E_F)}$ obtained with numerical simulations,[9] which are free from the MT approximations.

The present results should encourage the experimental obtention of magnetic multilayers with highly doped Si:P or similar systems as separators, which should show magnetic coupling even at extremely low temperatures. The limitation which our results impose upon the physically reasonable widths of such separators, namely, of the order of l, should in fact be relaxed considerably by a better theoretical treatment of the impurity-band susceptibility, which would avoid the averaging procedure of MT and the factorization of the product of Green's functions made in Eq. (28). We expect to find much longer localization lengths and, accordingly, a possibility of wider separators. Recent experiments have confirmed the feasibility of magnetic multilayers coupled through a semiconducting separator[10,11] at room temperature. The effective interaction we found can be applied, in principle, to the description of the magnetic properties of the impurity band at low temperatures in the metallic regime, where, as we mentioned before, a finite concentration of local spins persists.

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