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Bad-metallic behavior of doped Mott insulators

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Employing Nernst-Einstein decomposition $\sigma = e^2\chi_c D$ of the conductivity σ onto charge susceptibility (compressibility) χ_c and diffusion constant D, we argue that the bad-metallic behavior of σ in the regime of high temperatures and lightly doped insulator is dominated by the strong temperature and doping dependence of χ_c. In particular, we show how at small dopings χ_c strongly decreases towards undoped-insulating values with increasing temperature and discuss a simple picture leading to the linear-in-temperature resistivity with the prefactor increasing inversely with decreasing concentration (p) of doped holes, $\rho \propto T/p$. On the other hand, D shows weak temperature and doping dependence in the corresponding regime. We support our arguments by numerical results on the two-dimensional Hubbard model and discuss the proposed picture from the experimental point of view.

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Introduction. Many materials show puzzling metallic behavior in which the resistivity is linear in temperature up to a very high temperature, e.g., 1000 K, and smoothly crosses the Mott-Ioffe-Regel (MIR) limit without any indication [1]. Such behavior with resistivity being metallic (increasing with temperature) and larger than the MIR limit is termed the bad-metallic behavior, which is frequently associated also with the breakdown of the Fermi-liquid concept and the loss of coherent quasiparticles. Understanding this behavior is one of the central challenges of the solid state physics ever since its discovery [2] in the cuprate high-temperature superconductors, since it might be the key towards an understanding of the superconductivity. Bad-metallic behavior is observed also in many other classes of materials such as pnictides [3–5], fullerenes [6], vanadium dioxide [7], ruthenates [8], organic charge transfer salts [9], and nickelates [10].

Despite this ubiquitous behavior, consensus on its proper understanding is still missing. In the 1970s it has been discussed in terms of electron-phonon scattering [11,12], followed by discussion of strong electron-electron interactions and correlations [1,13,14], while more recent proposals include quantum criticality [15–17] and a bound on diffusion constant motivated by holographic duality [18]. It has been discussed also with approaches aiming at lower temperatures, e.g., marginal Fermi liquid [19], spin fluctuation [20,21], and charge density wave [22] scenarios.

We use the Nernst-Einstein relation

$$\sigma = e^2\chi_c D, \tag{1}$$

which relates conductivity σ to the charge susceptibility (compressibility) $\chi_c = \partial n/\partial\mu$ and diffusion constant D. e is electronic charge, n is electronic density, and μ is chemical potential. We argue that in doped insulators at high temperatures (T) the linear-in-T resistivity arises due to strong T dependence of $\chi_c \propto 1/T$ and is therefore a static effect, while the diffusion constant D has a rather weak T dependence. We also show that at small dopings (p) χ_c gets strongly suppressed with increasing T due to approaching insulating values, which strongly increases the resistivity $\rho = 1/\sigma$ and naturally leads to the resistivity crossing the MIR limit. We also show that χ_c has strong p dependence and discuss possible effects of antiferromagnetic (AFM) correlations.

Nernst-Einstein relation. Equation (1) deconstructs σ onto χ_c and D. χ_c measures the change of electronic density due to changes of μ and since μ plays essentially the same role as a static uniform electric potential $e\phi$, χ_c can be seen as the measure of electronic density redistribution in the presence of slowly varying external electric potential. A particularly interesting situation appears for doped antiferromagnetic Mott insulators, where in the insulator $\chi_c = 0$, while with doping a transition from an insulator to a metal appears with a discontinuous jump or divergence of n vs μ, i.e., $\chi_c = \infty$. These two extreme limits of χ_c very close in the phase diagram can lead to strong T and p dependence of χ_c and in turn of ρ, simply due to the T-broadening effect. On the other hand, if χ_c measures the tension for charge redistribution in the presence of an external potential, D describes the rate at which the electronic density redistributes and is therefore a dynamic quantity, e.g., it can be related to the mean square velocity $\langle v^2\rangle$ and the scattering rate $1/\tau$, $D = \langle v^2\rangle\tau/2$. We note that Eq. (1) can be derived on the operator level [23] and its validity is not limited to only certain phases, e.g., to Fermi liquids, and should be valid also in the incoherent bad-metallic regime [24].

Hubbard model results. To explore the behavior of χ_c and D of doped insulators we use the Hubbard model on a two-dimensional lattice, which allows the description of the insulating phase (with the possibility of AFM ordering) as well as of the metallic doped insulator. The model Hamiltonian is

$$H = -t\sum_{\langle i,j\rangle,s}(c^\dagger_{i,s}c_{j,s} + \text{H.c.}) + U\sum_i n_{i,\uparrow}n_{i,\downarrow} - \mu\sum_{i,s} n_{i,s}. \tag{2}$$

t is the hopping amplitude, $c_{i,s}$ ($c^\dagger_{i,s}$) is the annihilation (creation) operator of electron on a site i with spin s ($\uparrow$ or $\downarrow$), $\langle i,j\rangle$ denotes nearest-neighbor sites on a lattice (we focus only on a square and triangular lattice), U is the on-site Coulomb repulsion, and $n_{i,s} = c^\dagger_{i,s}c_{i,s}$. We evaluate quantities for this model by using the numerical finite-temperature Lanczos method (FTLM) [13,25] on finite clusters (16 sites), which allows a precise evaluation of physical properties at high T and is therefore suitable to tackle these questions. At low T finite-size effects appear [24]. We further set $\hbar = k_B = 1$.

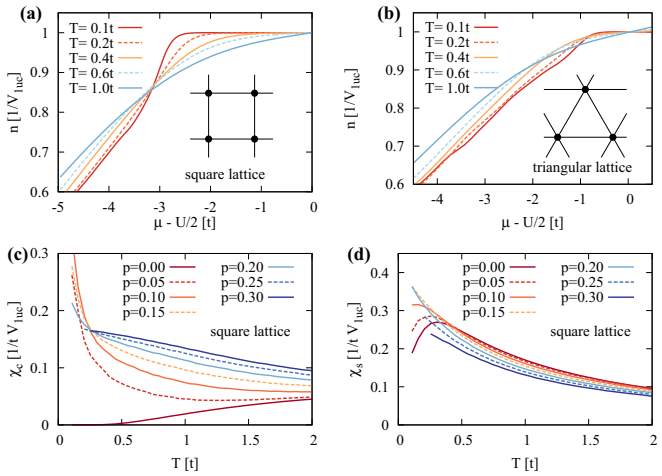

FIG. 1. Electron density n vs chemical potential μ and charge susceptibility ($\chi_c = \partial n/\partial\mu$) show strong T dependence at low dopings of Mott insulator. Results further suggest that χ_c is at low dopings increased by AFM correlations. (a) n vs μ for several T and square lattice, showing gapped behavior at $n = 1$ and finite slope at finite dopings ($n < 1$) with particularly large slope at small dopings. (b) n vs μ for several T and for triangular lattice, showing smaller slope at finite dopings than square lattice. This suggests χ_c at low dopings can be increased by AFM correlations, which are larger for a square lattice than for a frustrated triangular one. This is further supported by a comparison of panel (c), which shows χ_c vs T for several dopings and its increase at low T and low doping, with panel (d), which shows in the same regime suppressed uniform spin susceptibility χ_s. This suppression is due to stronger AFM correlations [27]. Results are calculated for $U = 10t$ and $V_{1\,\mathrm{uc}}$ denotes one-unit-cell volume.

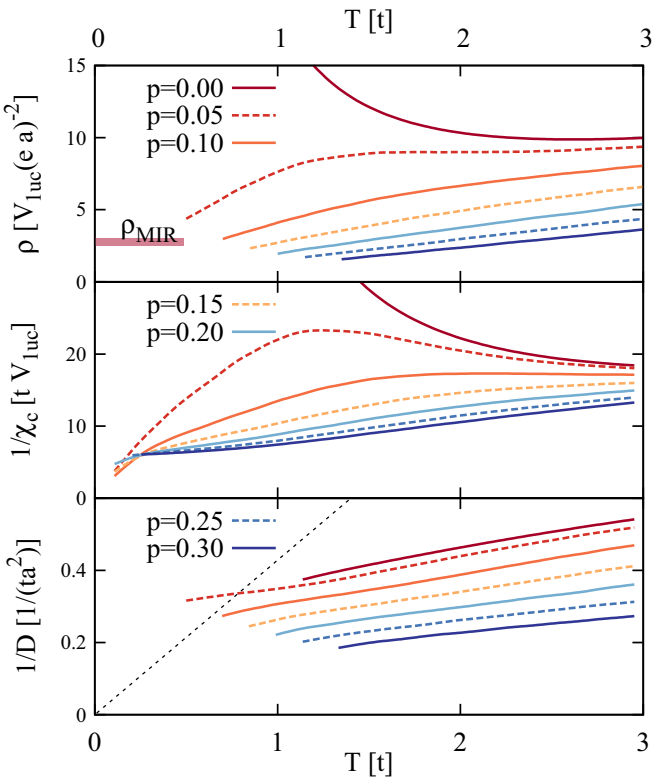

FIG. 2. Resistivity ρ (upper panel) of the doped Mott insulator shows strong T and doping ($p = 1 - n$) dependence at high T, and is dominated by the T and p dependence of inverse charge susceptibility $1/\chi_c$ (middle panel). $1/\chi_c$ clearly shows similar behavior as ρ. On the other hand, in the same regime diffusion constant (D) has a very weak T and p dependence. In particular, see the regime of $p = 0.05$ and $0.5t < T < t$ in which $\rho \propto T$ originates in $1/\chi_c \propto T$ and its increase by about a factor of 2, while D increases only by about 10%. The estimated upper bound on $1/D$ suggested by Hartnoll [18] is shown with a black dashed line in the lower panel. Results are calculated for a square lattice with $U = 10t$ and regimes potentially influenced by finite-size effects are not shown.

In Fig. 1(a) we show $n(\mu)$ calculated on a square lattice for several T and for $U = 10t$, which is within an insulating regime for half-filling ($n = 1$). At low $T \sim 0.1t$ a gapped behavior with $\chi_c = \partial n/\partial\mu = 0$ is observed at $n = 1$, while at finite hole dopings χ_c is finite. With increasing T the dependence of n on μ is smoothed. For $n = 1$ the behavior of χ_c is activated obtaining finite values at T comparable to the charge gap Δ_c [see Fig. 1(c) and Ref. [25]]. On the other hand, it is particularly evident that for small dopings ($n = 0.95$, $p = 1 - n = 0.05$) χ_c gets strongly reduced with increasing T and that μ is rapidly moved into the gapped regime for low T. This strong T dependence of χ_c is reflected in strong T dependence of ρ as shown below in Fig. 2. Such behavior originates in a simple electron number conservation, which makes the behavior of the doped system similar to the insulating one with increasing T [24]. We stress that this situation appears for any doped charge-gapped system, as long as the density of electrons is conserved (e.g., semiconductor, band insulator, Mott insulator, etc.).

χ_c of a lightly doped Mott insulator has additional features in contrast to, e.g., semiconductor or band insulators. At $T = 0$ and for $U \gg t$ the increase of U for δU leads to the increase of Δ_c at $n = 1$ to roughly $\Delta_c + \delta U$, but due to the same increase of $\mu_{\max}$ at which the band is completely filled ($n = 2$) to $\mu_{\max} + \delta U$, such increase of U does not (at least on average) change χ_c for finite dopings ($n \neq 1$) [24].

The effect of strong AFM correlations is different. The appearance of AFM correlations decreases the energy of the Mott-insulating state for an order of exchange energy J, makes it more stable, and increases Δ_c (see, e.g., Ref. [26] for indication of such behavior). For fixed U and in turn fixed $\mu_{\max}$ as well as $\mu_{\min}$, this needs to be accompanied with the increase of χ_c for $n \neq 1$. Such increase is seen in Figs. 1(a) and 1(c) at low dopings (e.g., $p \sim 0.05$) and at low T ($\sim 0.1t$), while it does not appear in a triangular lattice as shown in Fig. 1(b), since AFM correlations are decreased due to spin frustration. Our results in Fig. 1 are in agreement with results on a square lattice [13,27–30] showing (nearly) diverging χ_c and nondiverging χ_c on a triangular lattice [31]. Additionally the (spin frustrating) diagonal hopping reduces the divergence of χ_c and may explain differences in χ_c between cuprate families [32]. From this picture it is evident that AFM correlations can increase χ_c at finite dopings, which is discussed also by Imada [33] and indicated in the results of Ref. [34]. The increase of χ_c at low p and T due to AFM correlations is supported also by a simultaneous

decrease of uniform spin susceptibility χ_s in the same regime [see Figs. 1(c) and 1(d) and Supplemental Material [24]]. Furthermore, χ_c may also relate to the pseudogap [24], as is, e.g., discussed also by Sordi *et al.* [35–37].

In the following we show that strong p and T dependence of χ_c governs also $\rho = 1/\sigma$, as is expected from Eq. (1), in particular at high T and low p. This is the main result of this work. We first calculate via FTLM [13,25] optical conductivity $\sigma(\omega)$ as a dynamical current-current correlation function [24] and then extract dc conductivity $\sigma = \sigma(\omega \to 0)$. This method gives exact results for large $T > T_{\rm fs}$, while at low $T < T_{\rm fs}$ finite size effects appear. We do not show low T results which are potentially affected by finite size [24].

In Fig. 2 we show the T dependence of calculated ρ, $1/\chi_c$, and $1/D$, which we estimate from $1/D = \rho\chi_c/e^2$. It is clear that the T and p dependence of ρ is dominated by the T and p dependence of χ_c, while D shows a rather weak and modest dependence. The most intriguing dependence of ρ and $1/\chi_c$ appears in the low-T and low-p regime, where both quantities from small values at low T show strong and close to linear-in-T increase towards large insulating ($p = 0$) values. This originates in the move of μ into the charge gap with increasing T to conserve the density of electrons as discussed in the Supplemental Material [24]. On the other hand, $1/D$ shows a much more mundane dependence with weak increase with increasing T and small decrease with increasing p in the whole shown regime. For example, $1/\chi_c$ increases for $p = 0.05$ by about a factor of 2 from $T = 0.5t$ to $T = t$, while $1/D$ shows a small increase by about 10%. Similarly for $p = 0.05$, ρ shows a weak nonmonotonic T dependence, which becomes even more nonmonotonic for lower p, and originates in nonmonotonic T dependence of $1/\chi_c$. It cannot be understood via monotonic $1/D$. In the $T \to 0$ limit one expects the scattering rate and $1/D$ to go to 0 and therefore to strongly influence ρ. We do not reach this regime since finite-size effects appear first, but we stress that the system size dependence (not shown) of our results suggest weak T dependence of D and dominance of ρ by χ_c even for lower T than shown in Fig. 2 (see Supplemental Material [24] for more details). At lowest T and dopings ($p < 0.15$), $1/\chi_c$ shows additional suppression which presumably originates in the AFM correlations as discussed in Fig. 1, and could be related to the pseudogap.

From D we can estimate the mean free path l via $D = \langle v\rangle l/2$. The estimate of average velocity $\langle v\rangle$ is not straightforward and we use $\langle v\rangle = 8ta/\pi$, which together with the values of D from Fig. 2 gives l of the order of lattice spacing, $l \sim a$. Solely from l one would therefore not expect the crossing of the MIR limit [1] $\rho_{\rm MIR} = \sqrt{2\pi}V_{\rm luc}/(e^2a^2\sqrt{1-p})$. However, ρ does cross $\rho_{\rm MIR}$ due to a strong increase of $1/\chi_c$ towards the insulating values with increasing T as shown in Fig. 2. The crossing of $\rho_{\rm MIR}$ without any change or indication has therefore a natural explanation in our picture in terms of decreasing χ_c. And further, since there is no limit for $1/\chi_c$ close to the insulating regime ($1/\chi_c = \infty$), one does not expect an upper limit for the resistivity.

Hartnoll suggested [18] that the diffusion is bounded with the upper limit for $1/D < k_BT/(\hbar v_F^2)$. Here v_F is the Fermi velocity and by taking its bare band estimate $v_F \sim 8ta/\pi$ the calculated values of $1/D$ shown in Fig. 2 strongly violate the upper bound. However, by using the renormalized value $v_F \sim Z8ta/\pi$, with estimated renormalization $Z = 0.6$ [24] and showing the obtained upper bound with a black dashed line in Fig. 2, the violation region moves to lower T and becomes less apparent. Still this indicates a possible violation of the diffusivity bound at low T, but for a strict test v_F should be calculated separately [24].

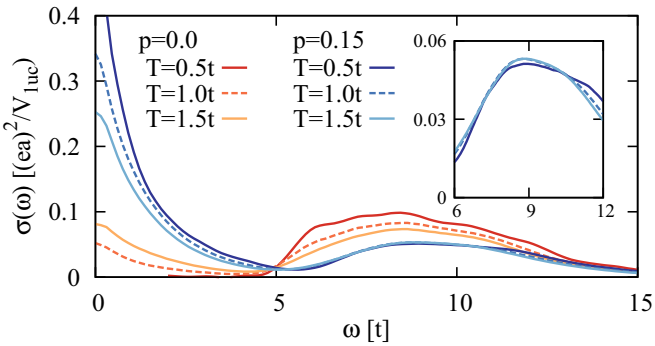

FIG. 3. Real part of conductivity $\sigma(\omega)$ for two dopings ($p = 0$ and $p = 0.15$) and for several high T ($0.5t$, t, and $1.5t$). The $p = 0.15$ case shows spectral weight transfer from low ω to high ω (see inset) with increasing T, and is simultaneously becoming more similar to the insulating case. The $p = 0$ case shows an opposite transfer of weight from high ω to low ω (activated behavior) with increasing T. Results are for a square lattice and $U = 10t$.

The strong T and p dependence of χ_c shown in Fig. 2 appears for any doped insulator, including Mott insulator, band insulator, and semiconductor, as long as n is fixed and not μ. In doped semiconductors the behavior is affected also by, e.g., acceptor levels ϵ_a above the valence band ϵ_v, which are localized ($D = 0$) [24]. However, the regime of small dopings and $\epsilon_v - \epsilon_a \ll T < E_g$, for which the chemical potential is in the charge gap E_g, has a textbook [38] density of conducting valence band holes $p_v = P_v e^{(\epsilon_v-\mu)/T}$. A similar relation between p, T, and μ is expected also in doped Mott insulators and has indeed been proposed also for cuprates [27,39] and readily leads to $1/\chi_c = T/p$. We further suggest this is the origin of $\rho \propto T/p$ at high T. We, however, stress that our results are only close to this picture since hole doping shifts μ into the lower band, while in semiconductors it stays above the valence band maximum.

Another feature arising naturally in the proposed picture is the optical spectral weight transfer, namely, the decrease of $\sigma(\omega)$ at low ω and increase of $\sigma(\omega)$ at higher ω with increasing T for finite doping and vice versa for $p = 0$. This is shown in Fig. 3 and is due to finite p behavior becoming more insulating like with increasing T. Simultaneously the spectra for $p = 0.15$ and $p = 0$ shown in Fig. 3 are getting similar at low and high ω. We note that some transfer can be suppressed by the decrease of the integrated spectral weight (absolute kinetic energy) with increasing T.

Experiments. It is of utmost importance to see whether experiments show any confirmation or indication of the proposed picture. We first need to determine the temperature denoted $T_{\rm coh}$, above which χ_c has strong T dependence. We estimate [24] $T_{\rm coh} \sim pW$ where W is half-bandwidth and $T_{\rm coh} \to 0$ as $p \to 0$. For example, for $t = 0.35$ eV and $p = 0.1$

we estimate T_{coh} of the order of 1000 K. However, for diverging χ_c at the MIT (see Figs. 1 and 2) T_{coh} can be reduced by an order of magnitude, e.g., to the order of 100 K for the chosen case. A similar estimate is discussed also by Imada [33].

By using photoemission spectroscopy (PES) it is possible to measure the T and p dependence of μ allowing for a direct experimental estimate of χ_c. The results for cuprates showing a strong increase of μ with increasing T already for $T \sim 200$ K, and a strong p dependence at small dopings [see Fig. 8.8(b) in Ref. [40]], suggest very large values of χ_c in the underdoped regime [41,42] and suggest strong and close to linear-in-T dependence of $1/\chi_c$ [43]. All these results are in qualitative agreement with our results and more importantly suggest that ρ can indeed be influenced by strong variations of χ_c already at quite low T. Unfortunately the direct comparison of experimental ρ and χ_c as well as extraction of D is unfeasible due to large uncertainties of PES data. Additionally other effects, e.g., Madelung potential [44,45], can potentially influence the PES data.

Since χ_c is a basic static quantity which measures the change of electronic density also due to the external electric potential, it influences also many other quantities. For example, Thomas-Fermi screening length of the external potential $l_{TF} \propto 1/\sqrt{\chi_c}$ [38] should have strong T and p dependence and the former is indeed supported by the experiment [46]. It is also expected that χ_c plays a considerable role for a phonon softening [47] and even for the superconductivity [30,33].

On the other hand, one can indirectly compare at least on a qualitative level the experimental high-T ρ and our $1/\chi_c$. Data on various cuprates [1,48–52] show $\rho \sim aT$ at high T with the prefactor a increasing with decreasing p in good qualitative agreement with our results in Fig. 2. A similar increase of the linear-in-T part of ρ has been observed also in the low-T regime [4,53,54], but we stress that at such low T $1/\tau$ and D are expected to give the dominant T dependence.

The behavior of ρ and χ_c shown in Fig. 2 is not expected just in the above discussed Mott insulators, but also for doped band insulators. Remarkably similar p and T dependence has been lately reported for the electrostatically doped band insulator MoS_2 [55]. Furthermore, in nickelates the bad-metallic behavior is related [10] to decreasing effective carrier density n_{eff} with increasing T while measured τ and effective mass showed rather weak T dependence. This is closely related to our decreasing-in-T χ_c and weak T dependence of D. Also the spectral weight transfer discussed above (see Fig. 3) is in qualitative agreement with optical conductivity for cuprates and many other bad-metallic materials [52,56–58].

Conclusions. We propose an explanation of large and linear-in-T resistivity of doped insulator in the bad-metallic regime in terms of a much simpler static quantity, namely, charge susceptibility χ_c. Furthermore, since χ_c should have similar T and doping dependence for any charge insulator (with the exception of insulators due to spatial localization of electrons) similar features in resistivity are expected for Mott and band insulators, semiconductors, as well as AFM slater insulators. We further discuss that in this simple picture the coefficient of linear-in-T resistivity is close to being proportional to $1/p$, while the diffusion constant D shows in the corresponding regime weak doping and T dependence. The same holds for the mean free path, which we estimate to a few lattice spacing. Crossing of the MIR limit observed in resistivity can be naturally understood as χ_c becoming small with increasing T due to the proximity of the insulating state. We note that our picture with weak T dependence of D is opposite to the main suggestion that $D \propto 1/T$ by Hartnoll [18] and is closer to the results by Pakhira and McKenzie [23], where D and its possible lower bound were discussed for a half-filled case [24] within a dynamical mean-field theory approach. Our data suggest a possible violation of the diffusivity bound [18], but a strict test would require a separate calculation of the Fermi velocity [24] which is beyond the scope of this work. Recently, the importance of χ_c for ρ was supported also with a high-T series expansion approach [24,59]. We further discuss how AFM correlations increase χ_c at low T, which might lead to pseudogap features in the resistivity.

Future challenges. It remains a theoretical and experimental challenge to establish in which T and doping regime the resistivity is dominated by χ_c and in which by D. A theoretical challenge is to reach lower T and in this respect continuous improvements of the insight into the phenomena, of analytical and numerical techniques as well as computational power is promising. We further comment in the Supplemental Material [24] on previous works [60–63], which indicate the importance of static effects.

Even more important is to experimentally establish the behavior of χ_c and D with T and p. In this respect PES, Thomas-Fermi screening length, and other possibilities are invaluable. D has been already measured in cuprates [64], but unfortunately for nonequilibrium quasiparticles. This makes it inapplicable to our analysis. Measurements of D are very challenging but also highly desirable. Experimental deconstruction of ρ onto χ_c and D would further pose strong constraints on the theories and would differentiate between them. They would, for example, offer insight on questions such as, is there a quantum critical point [15,16], is diffusion bounded as motivated by holographic duality [18], is there a regime with universal behavior of diffusion constant [65], is the pseudogap a new phase [66], is the nature of the insulator Mott, band, or (AFM) Slater-like. Deconstruction of ρ onto χ_c and D therefore offers an exciting, promising and yet to be fully explored approach towards a better understanding of the discussed phenomena, both from a theoretical and an experimental point of view.

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