Dominating Coulomb-interaction effects in amorphous In_xO_y films

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Insulating thin three-dimensional (3D) amorphous films of In_xO_y exhibit the surprising resistance-versus-temperature dependence $R \propto \exp[(T_0/T)^{1/2}]$ between 6 and 106 K. The $(1/T)^{1/2}$ exponent in the resistance expression, the observed positive magnetoresistance behavior, and the lack of dependence of the magnetoresistance upon magnetic field orientation all suggest a dominating Coulomb electron-electron interaction transport mechanism. Interestingly, the experimental magnetoconductance data could be well described using the 3D electron-electron spin-splitting interaction theory derived for metallic systems, but modified by a simple temperature prescaling factor, $1/\exp[(T_0/T)^{1/2}]$. This prescaling factor accounts for the strong temperature dependence of the conductivity that is present in insulating films.

In strongly localized films at low temperatures, the typical resistance between neighboring impurity sites often becomes larger than those resistances connecting some remote impurity sites. For a three-dimensional (3D) film having a constant density of states near the Fermi level the characteristic hopping length increases with lowering temperatures, leading to Mott's variable-range–hopping (VRH) law for the resistance[1]

$$\rho(T,\ B=0)=\rho_0\exp[(T_{0,\mathrm{Mott}}/T)^{1/4}]\ . \tag{1}$$

The magnetoresistance in the VRH regime can be positive (negative magnetoconductance) if a decrease in the overlap of the wave function "tails" occurs with magnetic fields, or the magnetoresistance can be negative (positive magnetoconductance) if quantum-interference effects dominate.[2–4] When Coulomb interactions are present between electrons, Knotek and Pollak showed that the density of states must have a minimum near the Fermi level.[5,6] Efros and Shklovskii showed that the single-particle density of states vanished at the Fermi level owing to the long-range nature of the Coulomb potential.[7] The dip between the filled and empty states has been termed the "Coulomb gap" (Δ), given by[8]

$$\Delta=e^3[N(E_F)]^{1/2}/(4\pi\epsilon\epsilon_0)^{3/2}\ , \tag{2}$$

where $N(E_F)$ is the unperturbed density of states evaluated at the Fermi energy level, ϵ is the relative permittivity or dielectric coefficient, and ϵ_0 is the free-space permittivity $=8.85\times10^{-12}$ C^2/J m. Pollak and also Hamilton considered the case when the density of states decreases according to a power law near the Fermi level.[9,10] For the case of a quadratic power-law dependence such that $N(E)\propto(E-E_F)^2$, the VRH law for the resistance takes on a new exponent; this Coulomb VRH dependence of the resistance becomes[8–10]

$$\rho(T,B=0)=\rho_0\exp[(T_0/T)^{1/2}]\ , \tag{3}$$

where T_0 is defined theoretically according to Shklovskii and Efros as[8]

$$T_0=2.8e^2/(k_B\xi 4\pi\epsilon\epsilon_0)\ . \tag{4}$$

In Eq. (4), ξ is the electron localization length whose magnitude is determined by knowledge of $T_{0,\mathrm{Mott}}$ in the Mott VRH regime and for a 3D film is given by[1]

$$\xi\simeq[k_BT_{0,\mathrm{Mott}}N(E_F)]^{-1/3}\ . \tag{5}$$

In order to observe the Coulomb VRH square-root power-law dependence of $\ln R$ on temperature, Shklovskii and Efros suggested that the experimental temperatures must be so low that the optimum Mott energy level band $E_{0,\mathrm{Mott}}$ becomes comparable to the Coulomb gap Δ,[8] namely,

$$E_{0,\mathrm{Mott}}\leq\Delta\ , \tag{6}$$

where

$$E_{0,\mathrm{Mott}}=k_BT(T_{0,\mathrm{Mott}}/T)^{1/4}$$
$$=(k_BT)^{3/4}/[N(E_F)\xi^3]^{1/4}\ . \tag{7}$$

The condition given by Eq. (6) suggests that Coulomb VRH dependence might be observable at temperatures below T_c where

$$T_c=T_0^2/T_{0,\mathrm{Mott}}=e^4N(E_F)\xi/[k_B(4\pi\epsilon\epsilon_0)^2]\ . \tag{8}$$

Entin-Wohlman *et al.* have derived the same results.[11] Alternatively, Castner, Shafarman, and Koon suggested that the condition to observe the square-root exponent of Eq. (3) is[12]

$$T\ll\Delta/k_B\ . \tag{9}$$

Do any of these predictions give reasonable values for the gap Δ,T_0 appearing in the exponent, and T_c the highest temperature at which the Coulomb VRH dependence might be expected? The problem in the evaluations is estimating reasonable values for the relative dielectric constant ϵ and the localization length ξ. Both of these parameters are enhanced due to the proximity of

the metal-insulator transition. From scaling-theory predictions,[13]

$$\xi(N)=\xi(0)(1-N/N_c)^{-\upsilon}$$

and (10)

$$\epsilon(N)=\epsilon(0)(1-N/N_c)^{-\zeta},$$

where $\xi(0)$ is typically on the order of the intercarrier distance $N^{-1/3}\simeq 50$ Å.[14] Experiments on the metal-insulator transition indicate that $\zeta/\upsilon\simeq 2$.[13] Ovadyahu has observed experimentally for In_2O_{3-x} films that $N(E_F)\simeq 10^{45}/\mathrm{J\,m^3}$, $\upsilon\simeq 0.8$, and $\xi(N)\simeq 200$ Å for our $\rho_{RT}=0.05\ \Omega\,\mathrm{cm}$ film.[14] For our case, the localization length is enhanced by a factor of 4, and hence, the relative dielectric constant ϵ should be enhanced by a factor of 16; since $\epsilon(0)\simeq 2$, then $\epsilon(N)$ for our case is approximately $\epsilon(N)\simeq 32$. Inserting these parameters into Eqs. (2), (4), and (8), we obtain reasonable values for Δ, T_0, and T_c; namely, $\Delta\simeq 6\times 10^{-22}$ J$=0.004$ eV$=44$ K, $T_0=73$ K, and $T_c=75$ K. These rough calculations along with the condition of Eq. (9) suggest that the Coulomb VRH dependence of Eq. (3) should easily be observed in indium oxide films.

Strongly localized, insulating, granular, transparent In_2O_{3-x} films have been studied rather extensively.[14–17] In the liquid-nitrogen to liquid-helium temperature range, these granular films exhibit the Mott VRH resistance dependence of Eq. (1) as well as negative magnetoresistances that are characteristic of the quantum-interference effects recently predicted by Sivan *et al.*[2,15,17] Only at liquid-helium temperatures where the hopping activation energies appear smaller than the Coulomb gap energy Δ does the R-versus-T dependence change over to the Coulomb VRH expression of Eq. (3); the magnetoresistance also takes on positive values.[17] Owing to the low-temperature limitations of 2 K of our cryostat, the Coulomb VRH properties could not be studied extensively in these granular In_2O_{3-x} films.

This article presents some new results on the electronic transport properties of amorphous opaque In_xO_y films. Ovadyahu has already observed Mott VRH dependences of the resistance of thick 1000–1500 Å amorphous In_xO_y films in the liquid-nitrogen to liquid-helium temperature range; he came to the conclusion that the Mott VRH properties are system independent.[14] Our results on thinner amorphous In_xO_y films probably challenge this claim—since we observed Coulomb VRH properties rather than the Mott VRH properties in our amorphous In_xO_y films at temperatures that are much higher than those temperatures observed in the granular films. We now present our results.

Amorphous In_xO_y films were prepared by thermally evaporating In_2O_3 powder from an alumina-coated Mo evaporation boat in a partial oxygen atmosphere of 5×10^{-5} mm Hg. The substrates were ordinary glass microscope slides maintained at room temperatures. A "cold" substrate is a necessary condition for obtaining the amorphous state.[18] Our resulting 460 Å thick amorphous In_xO_y films were quite opaque and brownish in color resulting from interband absorption. Our TEM results are very similar to those of thicker In_xO_y films reported in Ref. 14 in which very broad diffraction rings and a "sandy" featureless structure of the surface are present. These films could be easily and irreversibly converted into transparent granular In_2O_{3-x} films by heating them on a hot plate at 225° C in air for 10 min.

The low-temperature resistance versus temperature dependence for the amorphous film is shown in Fig. 1; over a wide temperature range of 6–106 K, the temperature dependence is characteristic of a Coulomb VRH process. The data can be well described by Eq. (3) with

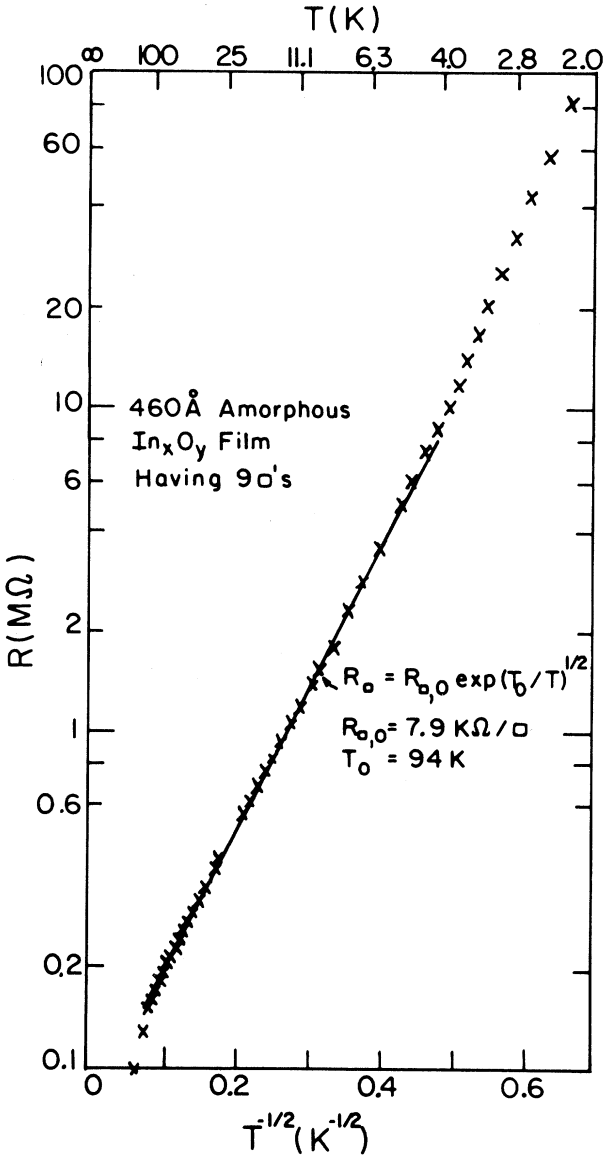

FIG. 1. Semilog plot of the resistance vs $T^{-1/2}$ for a 460 Å amorphous In_xO_y film. Note the large temperature range over which the Coulomb VRH dependence is observed. Conversion to resistivity ρ can be made using the relation $\rho=Rd/n_\square$, where d is the film thickness of 460 Å and $n_\square=9$ is the number of squares.

$T_0=94$ K and $\rho_0=0.0364$ Ω cm. The resistance data of Fig. 1 could not be fitted to a 3D Mott VRH expression of Eq. (1). In the liquid-helium temperature range, the resistance took on a stronger temperature dependence, where $R \propto \exp[(T^*/T)^p]$, with p being slightly less than 1. This new transport process has not be studied in detail by us.

The magnetoconductance (MC) was always negative above 6 K as shown in Fig. 2. The 3D magnetoconductance, $\Delta\sigma$, for a film of thickness d in cm is defined experimentally at a fixed measuring temperature as

$$\Delta\sigma[(\Omega\,\mathrm{cm})^{-1}]=1/\rho(B)-1/\rho(B=0)$$
$$=\frac{1}{d}[1/R_{\square}(B)-1/R_{\square}(B=0)]\ . \quad (11)$$

The negative sign of the MC data in Fig. 2 is a characteristic signature of electron-electron interactions in weakly localized metallic films. Notice in Fig. 2 that the MC data for this insulating film slowly decrease in magnitude as the temperature is lowered, which is in contrast to the behavior of the MC for a metallic film where the MC is predicted to increase to large negative values as the temperature is lowered.[19] Another signature for the spin-splitting electron-electron interaction is that the magnetoconductance should be independent of magnetic field orientation. Within our experimental error and reproducibility of ±25%, the MC in fields perpendicular to the film had the same values as the MC in fields parallel to the film.

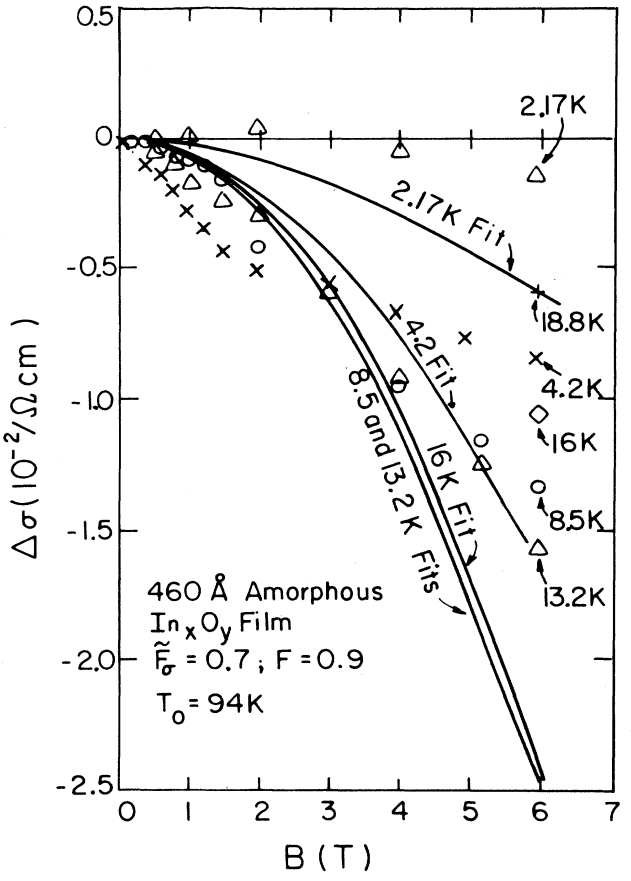

FIG. 2. Magnetoconductance data as a function of magnetic field at fixed, different temperatures. Most of the temperatures fall in the region where the Coulomb VRH dependence in the resistance is observed. The solid lines are fits of Eq. (18) using no adjustable parameters.

Is it then possible to describe the experimental MC results of Fig. 2 on this insulating film by using the electron-electron spin-splitting theory that was developed for a metallic film? Our observations suggest that it is possible to fit the theory to the data within a factor of 2 without the use of a single adjustable fitting parameter. We first point out that the electronic screening length l_s is on the order of 1.6 Å, which is one to two orders of magnitude smaller than the typical localization length ξ of the electrons in In_xO_y; hence, electron-electron interactions should always be present since the electrons are relatively mobile to interact and to screen one another. However, there is one important difference between the insulating and metallic films. The resistance of an insulating film can easily change by a factor of 10 or more over a decade change in temperature; the resistance change is particularly strong in the Coulomb VRH regime. In contrast, the change of resistance of a metallic film is typically a few percent over the same temperature interval. However, the metallic theory does not include the strong temperature dependence of the conductivity. We suggest that a multiplicative "prefactor" of the form $1/\exp[(T_0/T)^{1/2}]$ be included in the theoretical expression for the MC. This term, $1/\exp[(T_0/T)^{1/2}]$ has four desirable features. (i) As the film approaches the metal-insulator transition from the insulating side, one anticipates that $T_0\rightarrow 0$, since $\xi\rightarrow\infty$ at the transition; thus, the prefactor term takes on the value of unity, and the conventional electron-electron interaction expression for the MC is recovered for metallic films. (ii) This term gives a correct temperature-weighing factor that counteracts the increasing magnitudes of the MC $\propto T^{-3/2}$ which the theory predicts at lower temperatures. (iii) This term is unitless, and therefore no additional constants or parameters are required to cancel out the presence of any undersirable units. (iv) The only parameter appearing in this term (T_0) is easily and directly obtained from the R-versus-T data of the film.

We now review the 3D interaction formulas. There are two electron-electron contributions. One comes from an orbital effect of the magnetic field and has been discussed by Larkin, Altshuler *et al.*, and by Fukuyama.[20–22] The prefactor of the orbital term scales with the superconducting transition temperature of the film. Our In_xO_y films were insulating; hence $T_c=0$ and this orbital term was negligible.

The second interaction contribution, arising from spin splitting of the conduction electron energies, plays the dominant role in the transport properties of In_xO_y. The theory has been developed by Altshuler *et al.*;[23,24] the most recent developments appear in the review paper of Lee and Ramakrishnan.[25] According to Refs. 23, 24, and 25, the magnetoconductance $\Delta\sigma$ in the 3D interaction theory is negative:

$$\Delta\sigma[(\Omega\,\mathrm{m})^{-1}]=-\frac{e^2}{2\pi^2\hbar}\frac{\tilde{F}_\sigma}{2}[k_BT/(2D\hbar)]^{1/2}g_3(h)\ , \quad (12)$$

where D is the diffusion constant in units of m^2/s, $\tilde{F}_\sigma$ is the experimental electron screening constant, and $h=g\mu_BB/k_BT$. The asymptotic forms of $g_3(h)$ are[25]

$$g_3(h)=0.053h^2 \quad \text{for } h\ll 1\ ,$$

and (13)

$$g_3(h)=h^{1/2}-1.3 \quad \text{for } h\gg 1\ .$$

Our experimental values for h ranged between $\frac{1}{2}$ and 10, for which we used the simple and convenient series approximations for $g_3(h)$ derived by Ousset *et al.*[26]

$\tilde{F}_\sigma$ is related to the theoretical screening constant F through

$$\tilde{F}_\sigma=-\tfrac{32}{3}[1+0.75F-(1+F/2)^{1.5}]F\ . \quad (14)$$

Moreover, one can estimate F through knowledge of the Hall constant $R_H=V_Hd/IB$ by measuring the Hall voltage V_H and by using the theoretical expression derived by Lee for F (Ref. 27)

$$F=(2k_Fl_s)^{-2}\ln[1+(2k_Fl_s)^2]\ , \quad (15)$$

where $k_F=(3\pi^2/eR_H)^{1/3}$ is the Fermi wave vector and l_s is the electron screening length as defined by Ashcroft and Mermin[28]

$$l_s=(\pi a_0/4k_F)^{1/2}\ , \quad (16)$$

where a_0 is the Bohr radius ($a_0=0.53$ Å). From our measurements of the Hall constant $R_H=3.7\times10^{-8}$ m^3/C, we calculate the electron screening length to be 1.6 Å and the theoretical electron screening constant F to be 0.88. Using Eq. (14) $\tilde{F}_\sigma$ takes on the value of 0.64. The diffusion constant D can be calculated from

$$D(m^2/s)=1.52\sigma_{RT}(eR_H)^{1/3}\ . \quad (17)$$

Because the room-temperature conductivity σ_{RT} was small and on the order of $1950/\Omega\,m$, the diffusion constant took on a very small value of 5.3×10^{-6} m^2/s. Thus, both parameters of the theory $\tilde{F}_\sigma$ and D were known for our film.

Finally, the prescaling factor was inserted into Eq. (12) yielding the finalized expression for the MC

$$\Delta\sigma[(\Omega\,\mathrm{m})^{-1}]=-\frac{e^2}{2\pi^2\hbar}\frac{\tilde{F}_\sigma}{2}[k_BT/(2D\hbar)]^{1/2}g_3(h)\times\frac{1}{\exp[(T_0/T)^{1/2}]}\ . \quad (18)$$

Note that the MC values should be converted into units of $(\Omega\,cm^{-1})$. For small fields B and high temperatures T such that $h\ll1$, the MC expression takes on the following temperature and field dependences:

$$\Delta\sigma\propto B^2/\{T^{3/2}\exp[(T_0/T)^{1/2}]\}\ . \quad (19)$$

Surprisingly, identical dependences appear in the Coulomb VRH derivation to be discussed shortly.

The solid lines appearing in Fig. 2 are fits of Eq. (18) to the MC data using the predicted value $\tilde{F}_\sigma=0.7$. Within a factor of 2, the theory describes the experimental results. But owing to the weak temperature dependence of both the data and theoretical predictions, the MC curves for fixed different temperatures tend to lie close to one another making Fig. 2 difficult to comprehend. A much clearer presentation is made by plotting the MC values as a function of temperature for a fixed magnetic field; the MC data taken at the fixed field of 6 T appears in Fig. 3 along with the theoretical curve. In this case, the electron screening constant $\tilde{F}_\sigma$ was treated as an adjustable parameter to give the best fit to the MC data point at 8.6 K. In this case $\tilde{F}_\sigma$ had the value of 0.37, yielding a value of 0.75 for the theoretical screening constant F. This value is reasonably close to the theoretical predicted value of 0.88. Needless to say, we are surprised that this phenomenological theory describes the MC data so well.

An alternative explanation for the MC data is given by Shklovskii and Efros based upon the distortion (elongation and shrinkage) of the wave functions with magnetic field.[8] Note, however, that our data does not exhibit the gigantic positive magnetoresistance observed in many semiconductor materials. In the simplest case, the spherical wave function in the absence of a magnetic field becomes a cigar-shaped figure in the presence of a magnetic field. This leads to a sharp decrease of the overlap of the wave function "tails" for an average pair of neighboring impurities, and hence to an exponentially increasing resistivity. Of special interest is the situation where the density of states is governed by the Coulomb gap. For weak magnetic fields, Shklovskii and Efros find that[8]

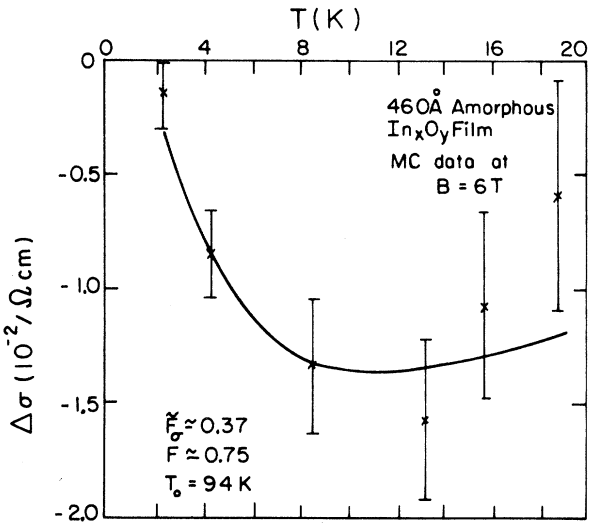

FIG. 3. Magnetoconductance data at the fixed magnetic field of 6 T as a function of temperature. In the fitting procedure $\tilde{F}_\sigma$ was treated as an adjustable parameter to force the theory through the 8.6 K data point. The resulting theoretical value for the electron screening constant $F=0.75$ was close to the theoretically predicted value of 0.88.

$$\rho(T,B)=\rho(T,B=0)\exp[0.0015(\xi/\lambda)^4(T_0/T)^{3/2}] , \quad (20)$$

where $\rho(T, B=0)$ is the zero-field resistivity given by Eq. (3), ξ is the localization length given by Eq. (5), and $\lambda=(\hbar/eB)^{1/2}$ is the magnetic length. Expanding the exponential $[\exp(x)\simeq 1+x]$ for small x's and hence small fields and high temperatures, we find for the MC

$$\Delta\sigma=-[0.0015(\xi/\lambda)^4(T_0/T)^{3/2}]/\rho(T, B=0) ,$$

or (21)

$$\Delta\sigma=-\frac{1}{\rho_0}(0.0015\xi^4e^2T_0^{3/2}/\hbar^2) \times B^2/\{T^{3/2}\exp[(T_0/T)^{1/2}]\} .$$

For reasonable values for the localization length $\xi\simeq 200$ Å, $T_0=94$ K, and $\rho_0=0.0363$ Ω cm from Fig. 1, the values predicted by Eq. (21) are within a factor of 3 compared to the experimental results. Equation (21) has the same field and temperature dependences as Eq. (19). Interestingly, Schoepe has observed these dependences experimentally in a series of elegant transport measurements made on a doped Ge sample.[13]

In conclusion, we have measured several amorphous indium oxide films of 460 Å thickness. These films exhibited Coulomb variable-range–hopping properties that were reproducible from run to run with an accuracy of ±25%. The reported results by Ovadyahu differ qualitatively from our results on amorphous In_xO_y films with very similar TEM pictures;[14] perhaps, this difference can be attributed to the thicker films that Ovadyahu studied or to substrate temperatures that were not maintained at room temperature during the long evaporations. Certainly, additional measurements are needed to clarify the electronic transport properties of amorphous In_xO_y having different thicknesses.

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