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The power law time dependence of $\langle R^2 \rangle$ in non-equilibrium growth

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Abstract. Model systems evolving in time to attain equilibrium, with spin exchange dynamics, order by forming domains after a temperature quench within the ordered region. They are described in terms of time-dependent diffusion coefficients and a distribution of activation energies. This description is analogous to the relaxation in disordered systems such as a:Si-H (amorphous hydrogenated Si).

Diffusive motion in solids [1], described usually in terms of a particle executing a random walk in a periodic potential, is characterized by the mean square displacement, $\langle R^2 \rangle$, that grows linearly with time. The temperature dependence of the diffusion coefficient has an activated form, with the activation energy the depth of the potential well. For systems with a finite number of particles and no interactions (except site exclusion where two particles cannot occupy the same site simultaneously), $\langle R^2 \rangle$ still grows linearly with time. The diffusion coefficient has the same activation energy but the prefactor is modified by the so-called lattice correlation factor, which expresses the tendency of a particle to jump into the hole left behind by a neighbouring particle that has already jumped. It only depends on the lattice geometry and the number of particles present. In this case, the mean square displacement is defined as

$$\langle R^2 \rangle = \frac{1}{N} \sum_{i=1}^{i=N} |R_i(t) - R_i(0)|^2 \quad (1)$$

where $i = 1, N$ goes over all the particles in the system and $R_i(t)$ is the displacement of the i th particle at time t . The correlation factors have been calculated for several lattices both with Monte Carlo simulations and analytically within the mean-field approximation.

For systems that have both a finite number of particles and interactions between the particles, diffusion becomes a non-linear process since the effective diffusion barrier of a particle depends on its local environment. After the particle jumps, it modifies the local environment, which in turn changes the probability to overcome the new barrier. It follows then that the diffusive behaviour depends on the initial configuration of the system, since it will produce different overlayer morphologies. For most well-equilibrated systems, $\langle R^2 \rangle$ still follows a linear time dependence, with the prefactor modified by the correlation factor described above, and a different activation energy, which is a result of sampling different local diffusion environments. Sublinear diffusive behaviour has been observed with Monte Carlo simulations [2], in systems that develop overlayer structures which contain 1D 'corridors'. It is known that blocking effects in 1D diffusion slow down $\langle R^2 \rangle$ because there are no sideways paths to bypass the blocking particle.

$\langle R^2 \rangle$ has not been measured systematically for a system under non-equilibrium conditions while the system is evolving in time, from an initial to a final state after a temperature quench. Although the growth of ordered domains has been extensively studied [3] and growth laws have been identified for the average domain size, it is still an open question to determine the time dependence of $\langle R^2 \rangle$. Only recently it was suggested [4] that $\langle R^2 \rangle$ grows sublinearly with time $\langle R^2 \rangle \sim t^{1-x}$ with x related to the growth exponent describing the growth of the average domain size L . As the ordered domains grow there are two kinds of diffusive atoms, the ones inside the domains which are in general more strongly bound and the ones at the domain perimeter which are weakly bound, because of the number of 'wrong' bonds at the boundary. At early time, when the domains are small, most of the atoms are at the boundary and because $E_{\text{out}}/kT < E_{\text{in}}/kT$, (where E_{out} , E_{in} are the diffusive barriers for 'outside' and 'inside' atoms respectively) one can assume that only the 'outside' atoms N_{out} contribute to $\langle R^2 \rangle$, so the contribution of the 'inside' atoms N_{in} can be neglected.

$$d\langle R^2 \rangle/dt = f_{\text{out}}N_{\text{out}} \exp(-E_{\text{out}}/kT) + f_{\text{in}}N_{\text{in}} \exp(-E_{\text{in}}/kT) \simeq f_{\text{out}}N_{\text{out}} \exp(-E_{\text{out}}/kT) \quad (2)$$

where f_{out} , f_{in} are the corresponding vacancy factors for the 'outside' and 'inside' atoms respectively, i.e., the fraction of nearest-neighbour vacant sites where a particle can jump into. Since N_{out} decreases as the perimeter/area of the growing domains, it follows that the rate of change of $\langle R^2 \rangle$ is proportional to the inverse of L . It is well documented [3] that L obeys a power law $L = A(T)t^x$, with x a universal growth exponent, i.e., it depends only on a few basic parameters and not the specific details of the system, and $A(T)$ the growth rate activation energy, which is a function of the temperature T . It can be easily seen that $L \sim t^x$ implies $\langle R^2 \rangle \sim t^{1-x}$.

It is important to emphasize that the above approximation depends on neglecting the second term in the RHS of (2). This approximation is better at lower temperatures (which is surprising because most traditional growth measures 'freeze' as the temperature is lowered), it depends on the density of the growing phases (with dense phases being more suitable for the approximation since $f_{\text{out}} \gg f_{\text{in}}$), it improves with coverage θ (since more particles are used for the averaging in (1)), but is expected to fail at long times when the two terms in (2) become comparable. So when the domains are sufficiently developed and equilibrium is being approached, which is the regime where most other growth measures are expected to reach their asymptotic time dependence, $\langle R^2 \rangle$ most likely will obey the linear equilibrium dependence, unless other topological constraints [2] (like the 1D behaviour observed in 'corridor' structures) is present.

Similar sublinear time dependence in $\langle R^2 \rangle$ have been observed [5] in a:Si-H and it is interesting to ask whether there are any analogies between the physical mechanisms responsible for the slower rates, observed in these two physically dissimilar situations. Experimentally, in a:Si-H the mean square displacement of the diffusing species grows as $\langle R^2 \rangle \sim t^{1-\alpha}$. This is equivalent to the time-dependent diffusion coefficient, $D \sim t^{-\alpha}$, which can be explained if a time-dependent vibrational rate of the relaxation process, is assumed $\nu = D(t)/a^2$, (where a is the lattice constant) or in terms of an exponential distribution of activation energies. Sites with different diffusion barriers which result in different jumping probabilities control the motion. By realizing the stochastic nature of the energetics in the a:Si-H system and in non-equilibrium growth kinetics which leads to sublinear $\langle R^2 \rangle$, one should search for a deeper analogy between the two situations.

Figure 1 shows the mean square displacement $\log \langle R^2 \rangle$ versus $\log t$. The results are shown both for repulsive (figure 1(a)) and attractive (figure 1(b)) nearest-neighbour

interactions at various interaction strengths J/kT . The initial state of the system in all cases is the random, infinite temperature configuration. This is the well-known Ising model at coverage $\theta = 0.5$ (zero magnetization) and critical temperature $|J/kT_c| = 1.86$. Spin exchange (Kawasaki) dynamics, where a particle exchanges with a nearest-neighbour empty site, are applied. Sizes of 61×61 are used (with few runs on larger sizes 101×101 that do not show any size dependence) but with excellent statistical averaging over 50 independent samples. Periodic boundary conditions are employed. Excellent averaging on the order of 5×10^4 independent terms is used in (1), the combination of summing over both particles and samples. Two different Monte Carlo algorithms have been used to implement the jumping probabilities. The first one allows particle-hole exchange with a probability $\exp((E_f - E_i)/kT)$ where E_i and E_f are the initial and final energies before and after the exchange. This probability is commonly used in Monte Carlo simulations of domain growth kinetics and obeys detailed balance. We have also used a different probability $\exp(-E_i/kT)$, to have a successful jump, which takes into account only the energy of the initial site, it only obeys detailed balance on the average and it has been recently suggested [6] to describe the kinetics more faithfully. We only show the results for the probability $\exp(-E_i/kT)$ and the complete set of data for both probabilities can be found elsewhere [7]. The results for $\langle R^2 \rangle$ are similar for the two probabilities; the one based on the $\exp((E_f - E_i)/kT)$ probability is faster, with domains reaching a certain size for a smaller number of Monte Carlo steps but with more meandering in the domain walls.

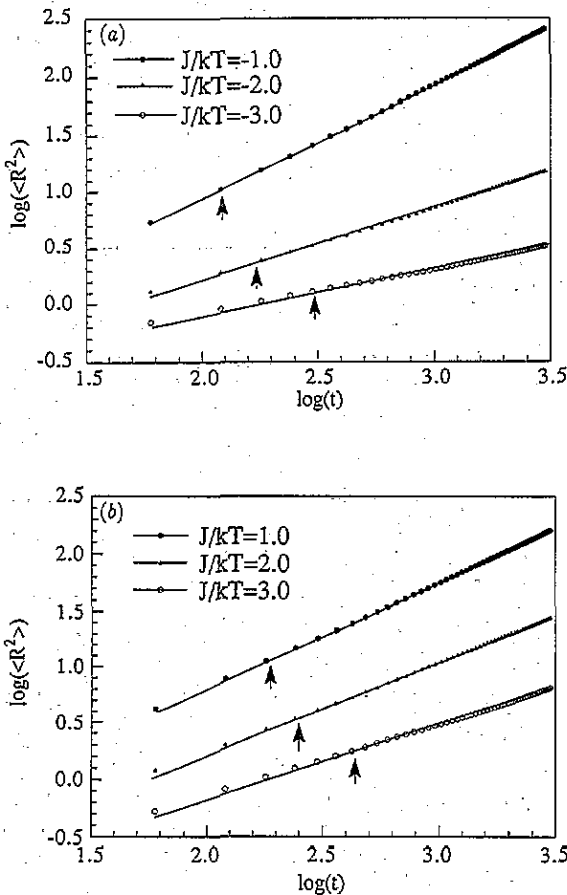


Figure 1. (a) Plot of $\log \langle R^2 \rangle$ versus $\log t$ for repulsive interactions $J/kT < 0$ at coverage $\theta = 0.5$. The value of the growth exponent is $x = 0.56 \pm 0.03$ for $J/kT = -3.00$ ($T/T_c = 0.62$), a quench deep into the ordered region where domains of the $c(2 \times 2)$ phase are formed. The range of data points used extends to later times denoted by the arrow. The results are shown for the single-site probability $\exp(-E_i/kT)$. Similar results are obtained for the standard algorithm $\exp((E_f - E_i)/kT)$. (b) Plot of $\log \langle R^2 \rangle$ versus $\log t$ for attractive interactions $J/kT > 0$ at coverage $\theta = 0.5$. The value of the growth exponent is $x = 0.32 \pm 0.01$ for $J/kT = 3.00$ ($T/T_c = 0.62$), a quench deep into the ordered region where domains of the (1×1) phase are formed. The range of data points used extends to later times denoted by the arrow. The results are shown for the single-site probability $\exp(-E_i/kT)$. Similar results are obtained for the standard algorithm $\exp((E_f - E_i)/kT)$.

Exponents can be obtained from the slopes of the data shown in figures 1(a) and 1(b). The results are summarized in table 1 for both sets of interactions and for the two algorithms. It is clear that for temperatures in the disordered region, for both types of interactions $|J/kT| = 1(T/T_c = 1.86)$ the effective exponent is $x \simeq 0$ which implies normal, linear diffusive motion as expected, because no domains are formed and no distinction between 'inside' and 'outside' atoms can be made. As the temperature is lowered at $|J/kT| = 2.00(T/T_c = 0.93)$ the exponent x becomes non-zero and when it is lowered further $|J/kT| = 3.0(T/T_c = 0.62)$, well within the ordered region, it increases to $x = 0.56 \pm 0.03$ for the case of repulsive interaction (and $c(2 \times 2)$ domains) but to a smaller value $x = 0.32 \pm 0.03$ for attractive interaction (and (1×1) ordered domains). The fit to obtain the above effective exponents is restricted to late times, marked by arrows in figures 1(a) and 1(b). If all the points were used then lower effective exponents by 10% were determined. It is expected, however, that early time data points include transients that should be avoided. Table 1 shows the value $\langle R^2 \rangle_{\max}$ at the end of the fit. It is clear that the sublinearity is not only temperature dependent (following the degree of order formed) but it is sensitive to the type of ordered structures formed (whether they are $c(2 \times 2)$ or (1×1)) and therefore the underlying growth mechanism.

Table 1. Listing of the growth exponents x extracted from $\langle R^2 \rangle \sim t^{1-x}$ for negative ($J/kT < 0$) and positive ($J/kT > 0$) interactions. Two different algorithms were used with probability of jumping $\exp((E_f - E_i)/kT)$ (detailed balance is satisfied) and $\exp(-E_i/kT)$ (detailed balance is satisfied on the average) where E_f, E_i are the final and initial energies used. $\langle R^2 \rangle_{\max}$ denotes the mean square displacement reached at the end of the growth. For negative interactions $\langle R^2 \rangle_{\max}$ is less and the average domain size $\langle L \rangle$ larger than for positive interactions, suggesting a different growth mechanism, that involves rearrangement of atoms at the boundary and no long range mass transport. $\langle L \rangle/N$ denotes the fractional average domain size reached, in agreement with results published in the literature [3, 8].

$J/kT < 0$							
$\exp(E_f - E_i/kT)$				$\exp(-E_i/kT)$			
J/kT	x	$\langle R^2 \rangle_{\max}$	$\langle L \rangle/N$	J/kT	x	$\langle R^2 \rangle_{\max}$	$\langle L \rangle/N$
-1.0	0.00	630	0.06	-1.0	0.00	250	0.05
-2.0	0.30	59	0.17	-2.0	0.35	16	0.22
-3.0	0.52	8	0.32	-3.0	0.56	3.5	0.25
$J/kT > 0$							
$\exp(E_f - E_i/kT)$				$\exp(-E_i/kT)$			
J/kT	x	$\langle R^2 \rangle_{\max}$	$\langle L \rangle/N$	J/kT	x	$\langle R^2 \rangle_{\max}$	$\langle L \rangle/N$
1.0	0.00	450	0.08	1.0	0.05	160	0.09
2.0	0.13	100	0.21	2.0	0.15	28	0.14
3.0	0.32	23	0.18	3.0	0.32	6.5	0.13

The values of the exponent x obtained from figure 1 are in remarkable agreement with the growth exponents deduced [3] from traditional growth measures like $S_{\max}(q, t)$, the maximum of the structure factor or the excess energy perimeter. It has been established analytically and with simulations that $x = \frac{1}{2}$ and $x = \frac{1}{3}$ are the corresponding growth exponents for repulsive and attractive interactions respectively, when the true growth regime is reached. Simulations sometimes have difficulty approaching the growth regime. The $x = \frac{1}{2}$ result has been easily obtained for spin-flip kinetics (non-diffusive evolution when the number of particles is not conserved) and with more computational effort for systems

with spin exchange dynamics. For the $x = \frac{1}{3}$ case, it has been difficult to reach the $t^{1/3}$ growth regime because of the conservation law. It is natural to ask whether we are indeed in the true growth regime, and if we are, why is $\langle R^2 \rangle$ so efficient to probe the growth law?

In addressing the first question we have monitored the increase in domain size with time, to ensure that $\langle R^2 \rangle$ is fitted to a power law during the same time interval that the domains have not stopped evolving. For negative interactions, we have calculated [7] the non-equilibrium structure factor at the Bragg peak $q_B = (\pi/a, \pi/a)$, $S(q_B, t)$ which grows like L^2 . $\langle R^2 \rangle$ is fitted during the increase of $S(q_B, t)$, well before the levelling off. Also, visual examination of the domain sizes shows that they are approximately 30% of the lattice size before finite size effects become important, as documented in other studies [3]. For attractive interactions, the full $S(q, t)$, at all points of the Brillouin zone, is needed to extract L which requires a larger computational effort. We rely on visual examinations of configurations printed out in our simulations, to compare them with published results in the literature, to test whether we have reached the same growth regime. We have used the 'chord length' as a measure of the domain size where the portions of a family of parallel lines, intercepted by the domains, were used to extract the average length. 'Chord lengths' up to 12% of the lattice size ($N = 61$) were measured, comparable to the sizes achieved in other published results [8], at the lowest temperature of the simulation $J/kT = 3.00$. The 'chord length' for the (1×1) structure is well defined because it is easily determined from the successive occupation of the same type of site (empty or full) to locate the end of the domains. In table 1 we also list the fractional domain size completed at the end of the regime, used for the fit, which also agrees with sizes obtained in the other studies [8]. We also list the final value of $\langle R^2 \rangle$ at the end of the fit for different temperatures. When diffusion is linear, very large increase of $\langle R^2 \rangle$ is attained. At low temperatures when domains are evolving, large sizes are attained for small $\langle R^2 \rangle$ displacements ($\langle R^2 \rangle = 8$ for $\langle L \rangle/N = 0.3$) when the interactions are repulsive. For attractive interactions, however, even after $\langle R^2 \rangle = 20$ domain sizes (as determined by the 'chord length' algorithm which overestimates the actual size) less than $\langle L \rangle/N = 0.12$ is completed. This confirms the underlying different mechanism [3] operating for the two types of interactions. For attractive interactions mass transport is necessary, in addition to the elimination of the domain boundary for the growth of the domains (which leads to slower growth $x = 1/3$, and larger $\langle R^2 \rangle$ displacement), while for repulsive interactions no mass transport is needed ($x = 1/2$ and $\langle R^2 \rangle$ is smaller).

Next we address the second question, that is, why $\langle R^2 \rangle$ is so efficient a growth measure. It has already been pointed out [9] that self-averaging growth measures, which improve statistically as the system size increases, are more reliable measures for extracting growth exponents. $\langle R^2 \rangle$ is such a self-averaging measure and its accuracy depends only on the difference in the energetics of the 'inside' to the 'outside' atoms. With increase lattice size and number of particles, this difference is becoming more pronounced and the approximation becomes statistically more accurate. The approximation improves at lower temperatures as pointed out before. In addition, the ferromagnetic ordered phase (1×1) formed for the case of attractive interactions, has a zero-vacancy factor for the 'inside' atoms, thus justifying the approximation at any temperature.

As stated before, the measured exponents, based on $\langle R^2 \rangle \sim t^{1-x}$ are obtained in the regime where domains are growing. Within this regime the exponents are constant and consistent with the values expected for the corresponding domain growth exponents. This holds until some characteristic time t_f , where the approximation applied to (2) is justified. After this time the two terms in (2) are comparable, and if effective components are extracted from $\langle R^2 \rangle \sim t^{1-x}$, they will be temperature dependent. In this crossover regime the 'inside' and 'outside' atoms are contributing equivalently, which can result in temperature-dependent

exponents until eventually in equilibrium the $x = 0$ value is recovered. In this linear regime a smaller slope of $D = (R^2)/4t$ is expected, since a higher diffusion activation energy controls equilibrium diffusion. Our simulations for the temperature $T < T_c$, are rather limited in the early-time, constant growth exponent regime $t < t_f$.

It is clear why the sublinearity obeyed by (R^2) in non-equilibrium systems and in a:Si-H has a common origin. One can identify a correspondence between the types of sites in the two systems. The deep sites in a:Si-H are equivalent to the 'inside' particles where once a particle is captured it is less mobile. 'Shallow' sites correspond to the 'outside' atoms, whose decreasing number with time is causing the sublinear dependence. In both cases one has a distribution of activation energies, for a:Si-H reflecting the local H configuration, and for non-equilibrium systems expressing the local environment of a diffusing atom. The exact type of energy distribution, whether it is exponential or not, does not affect the analogy.

It has been observed in all experimental domain growth kinetic studies that eventually growth levels off and the final size is less than the optimal achievable size thermodynamically. The levelling-off has been observed in both 2D and 3D systems, spanning a wide range of substrate-overlayer combinations. This growth limitation has been traditionally associated [3] with the presence of impurities that limit the further evolution of domains once they reached a certain size. However, if the experiment is continued over longer periods of time (measuring growth the next day), there is essentially no growth, well below the logarithmic prediction expected if impurities are responsible. This lack of growth has been observed [10] previously in the random-field Ising model system $Mn_{0.75}Zn_{0.25}F_2$ where domains also stop growing well below the expected logarithmic prediction, suggested theoretically.

In pure systems, theoretically it has been established that only finite size effects limit the growth. On the other hand, for a:Si-H, the time dependence of the 'deep' site occupation follows a stretched exponential, with long time saturating tails approaching the equilibrium value, reminiscent of the levelling-off observed in non-equilibrium systems. It is possible that at late stages of growth in non-equilibrium systems, the distribution of activation energies becomes analogous to the distribution of activation energies in a:Si-H system implying stretched-exponential relaxation. Theoretical model systems have a different distribution at late times which excludes saturation effects. This distribution of activation energies in non-equilibrium systems is time dependent while the distribution in a:Si-H is static. At very long times, the distribution in non-equilibrium systems should change to a single-valued distribution that corresponds to the uniform occupation of the surface with the ordered structure. It is possible that at this late time the experimental and model non-equilibrium systems behave differently.

A distribution of activation energies leads to the so-called Meyer-Neldel relation that has been observed [5] in the relaxation of the a:Si-H systems. It relates the prefactor $D_0(\omega)$ to the activation energy $E(\omega)$ of the diffusion coefficient measured at different frequencies ω (or equivalently, at different times)

$$D_0(\omega) = D_{00} \exp(E(\omega)/kT_0) \quad (3)$$

where D_{00} is some constant and T_0 is a transition temperature. Equation (3) follows from the progressive filling of the deep energy sites with time, that increases the activation energy and the number of occupants in the deep sites. The latter is proportional to $D_0(\omega)$. Equation (3) can be tested in ordering experiments, if measurements are performed at different times in the ordering process. For the O/W(110) system at high coverage ($\theta \simeq 0.6$), experiments were performed at the beginning of the ordering process [11] and at late times [12] when

equilibrium is reached. In the equilibrium experiment, $E^{\text{eq}} = 1$ eV and $D_0^{\text{eq}} = 10^{-4}$ cm² s⁻¹ are obtained. In the ordering, early-time experiments, $E^{\text{neq}} = 0.6$ eV is obtained. However, it is not yet clear how to extract the prefactor of the diffusion coefficient in non-equilibrium experiments as obtained from $A(T)$. Other unknown parameters such as the type of ordered domains formed and the interfacial tension relate $A(T)$ to the diffusion coefficient. However, it has been argued previously [13] that diffusion in the ordering experiment is equivalent to equilibrium diffusion at low coverage. Thus, if the low coverage values of $D_0^{\text{neq}} = 10^{-7}$ cm² s⁻¹ and activation energy $E_0^{\text{neq}} = 0.6$ eV are used for the non-equilibrium quantities [12] and $T_0 = 800$ K for the transition temperature when the system becomes disordered, then we can test whether these values are consistent with (3). We have

$$D^{\text{eq}}/D^{\text{neq}} = 10^3 \simeq \exp((E^{\text{eq}} - E^{\text{neq}})/kT_0) \quad (4)$$

suggesting that (3) is satisfied. Although this is only partial evidence that the Meyer-Neldel relation holds in non-equilibrium systems, the above discussion is suggestive of future experiments. The diffusion coefficients can be measured at various time intervals during the ordering process to test if (4) is verified.

It is interesting that the presence of many activation energies in a disordered system is a result of intrinsic disorder structurally related to the stoichiometry. However, for a quench system under non-equilibrium conditions with no disorder present, relaxation to equilibrium is controlled by the morphologically different local configurations that give rise to different local energy barriers.

The evidence presented above suggests strong, but incomplete, analogies between disordered and non-equilibrium systems. It is not clear yet what is the temperature dependence of the growth exponent x . For a:Si-H, it is expected [5] to show linear dependence with temperature, from $x = 0$ at zero temperature to $\beta = \frac{1}{2}$ at T_0 . However, different experimental studies [14] give widely different exponents for the same temperature range. It is known [3] from domain growth studies that defects and impurities can change drastically the values of the growth exponents so it is interesting to characterize the amount of impurities present in a:Si-H and correlate it to the varying exponents. In non-equilibrium systems, the exponent x is, in general [3] temperature independent unless the system is close to zero temperature or the critical temperature when the growth freezes. For the O/W(110) p(2 × 1) + p(2 × 2) system [11], it is $x = 0.31 \pm 0.03$. It is not clear what the physical origin is of the temperature dependence of x in a:Si-H and whether the different physical processes linked in this study, despite their unifying theme of multi-relaxation microscopic processes, have unique features that result in different temperature dependence for x .

Although there is strong evidence to relate the power law dependence of $\langle R^2 \rangle$ to the value of the growth exponent, the main point of this paper is independent of whether such a relationship exists: in both non-equilibrium and disordered systems $\langle R^2 \rangle$ grows with a sublinear time dependence. Additional analogies between the two phenomena are the distribution in activation energies, the existence of 'deep' and 'shallow' sites, the validity of the Meyer-Neldel relation and the slow, stretched-exponential relaxing observed asymptotically. The similarities, although incomplete, show promise to initiate further work on unifying these diverse time-dependent phenomena.

Acknowledgments

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