Submonolayer epitaxial growth with long-range (Lévy) diffusion

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The effects of long-range (Lévy) diffusion in submonolayer epitaxial growth are studied via kinetic Monte Carlo simulations and rate equations. Such long-range diffusion may be relevant in the case of liquid-phase epitaxy and electrochemical deposition. Results for the scaling of the submonolayer island density and size distribution are presented as a function of the Lévy distribution exponent β and the ratio D/F of the diffusion rate to the deposition rate. Both one- and two-dimensional Lévy flights (corresponding to infinitely fast hops) and one- and two-dimensional Lévy walks (corresponding to finite hopping velocity) are examined. Good agreement is found between theoretical predictions and simulations for the dependence of the island-density scaling exponent χ on the Lévy exponent β in both one and two dimensions. [S1063-651X(98)12012-3]

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I. INTRODUCTION

The growth of thin films by deposition techniques such as molecular beam epitaxy (MBE) involves nucleation, aggregation, and coalescence of islands on a two-dimensional substrate [1]. In the submonolayer regime this leads to the formation of islands of various sizes and morphologies, which grow and eventually coalesce to form a complete layer.

The standard theoretical approach to submonolayer growth [2-11] involves the use of rate equations [12] that describe the processes of adatom diffusion or hopping, island nucleation and growth, and deposition. For the simplest case (corresponding to a critical cluster size of 1 [2]) in which single adatoms may diffuse with hopping rate *D* while dimers and all larger clusters are stable and immobile, the standard rate-equation theory [2,4,5] predicts that the (per site) island density *N* at fixed coverage θ scales as

$$N \sim (D/F)^{-\chi},\tag{1}$$

where *F* is the (per site) deposition flux and $\chi = d/(2d+2)$ (where *d* is the dimensionality of the substrate). In particular, $\chi = 1/3$ for the case of deposition onto a two-dimensional substrate and $\chi = 1/4$ for the case of deposition onto a one-dimensional substrate or for deposition on a two-dimensional substrate with highly anisotropic diffusion. This type of scaling behavior for the island density has been verified in a large number of experimental and theoretical studies in which diffusion occurred via short-range (nearest-neighbor and next-nearest-neighbor) hops.

While in the case of thin-film deposition *in vacuo*, atoms are expected to diffuse via relatively short-range hops [13], in a number of experiments [14–16], long-range Lévy diffusion [17] of atoms at the liquid-solid interface has been observed. Such "long-range" diffusion may be relevant in thin-film growth by electrochemical deposition [18]. Therefore, the study of thin-film growth with long-range diffusion [14–21] may be of interest in connection with a variety of

experiments. From a theoretical point of view such a study is also of interest since it may lead to an improved understanding of nucleation and scaling in submonolayer growth.

In this paper we present both analytical (rate-equation) and simulation results for the scaling of the island density and island-size distribution for the case of submonolayer epitaxial growth with long-range (Lévy) diffusion [17]. The organization of this paper is as follows. In Sec. II we first review what is known about Lévy diffusion. In Sec. III we present a rate-equation theory for the scaling of the island density as a function of deposition rate in the case of long-range diffusion. In Sec. IV we describe in detail our simulations of deposition with Lévy diffusion. In Sec. V we present our simulation results for the scaling of the island density and island-size distribution for both Lévy walks and Lévy flights in one and two dimensions and compare them with our scaling theory. Finally, in Sec. VI we offer some conclusions.

II. LONG-RANGE (LÉVY) DIFFUSION

For ordinary Brownian diffusion, the mean-square displacement $\langle r^2(t) \rangle$ of a diffuser's position about its initial position is given by

$$\langle r^2(t) \rangle \sim (Dt)^{\mu},$$
 (2)

where $\mu = 1$ and *D* is the diffusion coefficient and *t* is time. The linear relationship between the mean-square displacement and time is a consequence of the finite second moment of the hop length along with the assumptions of translational invariance and absence of drift.

However, in the case of Lévy diffusion the secondmoment of the hop length is infinite. In particular, for the case of a constant-velocity Lévy walk in *d* dimensions with Lévy exponent β [which corresponds [19] to a probability density for the time spent in flight given by $\psi(t) \sim 1/(1 + t)^{d+\beta}$] the probability for a hop of length *x* is given for large *x* by

$$P(x) \sim x^{-d-\beta}.$$
 (3)

7130

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In this case, the mean-square displacement of a diffuser's position about its initial position is given by $\langle r^2(t) \rangle \sim (Dt)^{\mu}$ where

$$\mu = 1 \quad \text{for} \quad \beta \ge 2,$$

$$\mu = 3 - \beta \quad \text{for} \quad 1 \le \beta \le 2, \qquad (4)$$

$$\mu = 2 \quad \text{for} \quad \beta \le 1.$$

For $\beta > 2$, the second moment of the hop length is finite and one has ordinary Brownian diffusion. However, for $\beta \le 2$, the second moment diverges and one has anomalous diffusion. In particular, the region of β with $1 < \beta < 2$ is often referred to as enhanced diffusion while the range of β with $\beta < 1$ corresponds to "ballistic" diffusion.

In this study we consider submonolayer growth with two types of Lévy diffusion: Lévy "walks" and Lévy "flights." For both Lévy walks and flights with $\beta < 2$ the second moment of the monomer hop length is infinite. However, a Lévy walk assumes a finite hopping velocity whereas in our study the Lévy flight is an instantaneous jump where one jump is completed in one time step.

III. SCALING THEORY FOR χ WITH LONG-RANGE DIFFUSION

A simple scaling theory for the flux dependence of the island density at fixed coverage in one and two dimensions has been proposed in Refs. [3-5] for the case of short-range diffusion. Here we present similar arguments for the case of Lévy walks and flights in one and two dimensions.

For the case of submonolayer growth with irreversible attachment, the rate of change of the total island density N (corresponding to the rate of island formation) is equivalent to the rate at which dimers are formed when two monomers meet. This may be written as the product of the total decay rate of monomers $R_{\tau} = n/\tau$ (where *n* is the monomer density and τ is the monomer "lifetime") times the probability $P_1 = n/(N+n)$ that a monomer has collided with another monomer during this time rather than with an existing island. Thus, one may write

$$\frac{dN}{d\theta} = \left(\frac{n}{F\tau}\right) \left(\frac{n}{N+n}\right).$$
(5)

In the steady state the rate of deposition of monomers is exactly balanced by the rate of absorption due to encounters with other monomers and existing islands so that $n \sim F\tau$ while $n/(N+n) \approx n/N$ since $n \ll N$. Similarly, in the steady state the average distance $\langle r^2(\tau) \rangle^{1/2} \sim (D\tau)^{\mu/2}$ a monomer travels during its lifetime τ is of the order of the typical island distance $l \sim N^{-1/d}$ for d=1,2. Equating these two distances, one obtains $\tau \sim 1/(DN^{2/\mu d})$. Substituting into Eq. (5) and integrating (and ignoring the coverage dependence of N) one obtains,

$$N \sim (D/F)^{-\mu d/(2\mu d+2)}$$
 (6)

which implies that

$$\chi = \frac{d}{2d + 2/\mu}.\tag{7}$$

For the case of Brownian diffusion $\mu = 1$, so that Eq. (7) implies the standard result, $\chi = 1/3$ in d=2 and $\chi = 1/4$ in d=1 for this case. However, for the case of a Lévy walk with anomalous diffusion, one has $\mu = 3 - \beta$ for $1 < \beta < 2$ and $\mu = 2$ for $\beta < 1$. Thus, for a *d*-dimensional Lévy walk Eq. (7) implies

$$\chi = \frac{d}{2+2d}, \ \beta \ge 2, \tag{8a}$$

$$\chi = \frac{(3-\beta)d}{2+(6-2\beta)d}, \quad 1 \le \beta \le 2, \tag{8b}$$

$$\chi = \frac{d}{2d+1}, \quad \beta \leq 1.$$
 (8c)

This implies that for a one-dimensional Lévy walk (d=1),

$$\chi = 1/4, \quad \beta \ge 2, \tag{9a}$$

$$\chi = \frac{3 - \beta}{8 - 2\beta}, \quad 1 \le \beta \le 2, \tag{9b}$$

$$\chi = 1/3, \quad \beta < 1, \tag{9c}$$

while for a two-dimensional Lévy walk (d=2),

$$\chi = 1/3, \quad \beta \ge 2, \tag{10a}$$

$$\chi = \frac{3 - \beta}{7 - 2\beta}, \quad 1 \le \beta \le 2, \tag{10b}$$

$$\chi = 2/5, \quad \beta < 1.$$
 (10c)

Using similar reasoning, we can also obtain a rateequation prediction for $\chi(\beta)$ for the case of one- and twodimensional Lévy flights, for which the hop is taken to be instantaneous. In this case, one does not expect a significant difference in the value of the scaling exponent χ between Lévy walks and Lévy "flights" for $\beta > 2$ since the average hop length is close to 1. However, for $\beta \leq 2$ the average hop length increases with decreasing β (and becomes infinite for $\beta < 1$) so that a significant difference is expected. In particular, we assume that for Lévy flights rather than walks, and for $\beta < 2$, that a lower bound for the lifetime τ of a monomer corresponds to the elapsed time before a hop of length equal to the typical island distance $l \sim N^{-1/d}$. We expect that in d =1 this gives a relatively good estimate for the monomer lifetime (since a monomer which has jumped a typical island distance will almost certainly collide with an island) while in d=2 this only gives a relatively weak lower bound.

From Eq. (3) (with d set equal to 1 even in two dimensions since our two-dimensional simulations corresponded to independent one-dimensional hops in the x and y directions) the probability of such a hop may be written as

$$P(l > N^{-1/d}) \sim N^{\beta/d}.$$
(11)

If we now assume that the monomer lifetime τ is proportional to the inverse of this probability, i.e., $\tau \sim 1/(DN^{\beta/d})$, we obtain

$$\chi = \frac{d}{2+2d}, \quad \beta \ge 2, \tag{12a}$$

$$\chi = \frac{d}{\beta + 2d}, \quad \beta \leq 2, \tag{12b}$$

for the d-dimensional Lévy flights considered here [22].

IV. SIMULATIONS

In order to study submonolayer growth with Lévy diffusion we have carried out kinetic Monte Carlo simulations in one and two dimensions for the case of irreversible (nearestneighbor) attachment corresponding to a critical island size of 1. In particular, at each instant of time either a deposition or a diffusion move is selected according to the following probabilities:

$$p_F = \frac{1}{1 + N_1(D/F)}, \quad p_D = \frac{N_1(D/F)}{1 + N_1(D/F)},$$
 (13)

where p_F is the probability of selecting a deposition move and *F* is the (per site) deposition flux, p_D is the total probability of picking a diffusion move, and N_1 is the density (per site) of adatoms on the surface. If a deposition move is selected, then an adatom is deposited on a randomly selected site. If a diffusion move is selected, then an adatom (monomer) with no bonds is selected and allowed to jump (with a length given by the Lévy distribution) in a randomly chosen direction corresponding to one of the nearest-neighbor lattice directions.

In order to satisfy the probability distribution for Lévy flights given in Eq. (3), the jump lengths X were generated using the formula

$$X = [r^{-1/\beta}], \tag{14}$$

where *r* was a uniform random number between 0 and 1 and the brackets denote the closest integer. It is easy to show that in this case Eq. (14) implies that $P(X) \sim X^{-(\beta+1)}$ for large *X*, while the minimum hop length is equal to 1. We note that since in our two-dimensional simulations the hopping directions were restricted to the four nearest-neighbor directions of a square lattice, in this case the resulting Lévy flights actually corresponded to two independent one-dimensional Lévy flights/walks rather than the usual two-dimensional Lévy flight for which the direction chosen is completely random and continuous. Accordingly the use of Eq. 14 (without any modification for substrate dimensionality) in both one and two dimensions led to the appropriate scaling of the form of Eq. (4) for the mean-square displacement.

In our simulations four different cases were studied: onedimensional Lévy walks, one-dimensional Lévy flights, twodimensional Lévy walks, and two-dimensional Lévy flights. In the case of the one-dimensional Lévy flight a diffusing monomer was allowed to jump up to a distance X in one diffusion step (where $X \ge 1$) but must first visit all sites in between. By visiting all interim lattice sites, the flight may



be terminated short of its designated total hop distance due to an existing island or monomer occupying a site in the diffuser's path or nearest neighbor to the path.

For the Lévy walks, the situation was similar to that for Lévy flights. However, in this case the adatoms are assumed to diffuse with a finite velocity that allows for only one hop for each diffusion step. Accordingly, a list of each diffusing adatom's direction of jump and distance left to jump was kept along with the diffuser list. As in the flight case, in the case of Lévy walks the diffuser is capable of being stopped short of its jump length if it encounters other adatoms or islands along the way. In this case it is removed from the monomer list and either creates an island by nucleation with another monomer or is added to an existing island.

For one-dimensional walks and flights, the system size L was varied from 80 000 to 100 000, while for twodimensional walks and flights L varied from 300 to 1000. In particular the larger values of L were used for the case of small β in two dimensions in order to avoid finite-size effects due to the large hopping length. Averages were taken over of the order of 30 runs. For each run, data was collected for twenty coverages ranging from $\theta = 0.04$ to 0.8. The parameter D/F corresponding to the ratio of the diffusion (hop) rate to the deposition flux ranged from $D/F = 10^5$ to 10^9 . These are typical values for molecular beam epitaxy.

V. RESULTS

A. One-dimensional Lévy walks

Figure 1 shows typical simulation results for the dependence of the island density *N* as a function of the coverage θ for values of the Lévy exponent β ranging from 0.67 (ballistic regime) to 4 (Brownian diffusion) [23] for the case of deposition with diffusion via a one-dimensional Lévy walk. As shown in Fig. 1, the overall island density decreases with decreasing β due to the increased hopping length. Similarly, the range of coverage over which the island density is approximately constant appears to be increasing with decreas-





FIG. 2. Maximum island density N_{max} as a function of D/F for deposition with diffusion via one-dimensional Lévy walks for same values of β as in Fig. 1. Dashed lines correspond to power-law fits.

ing β . This behavior is similar to what has previously been observed [8] for normal diffusion as D/F is increased. Similarly, Fig. 2 shows typical results for the dependence of the peak island-density on the ratio D/F of the diffusion to the deposition rate for the same values of β as in Fig. 1, along with power-law fits to determine χ . As expected, for $\beta > 2$ the value of χ is close to 1/4 while for $\beta < 2$ the value of χ is larger than 1/4.

Figure 3 summarizes our simulation results for the dependence of the scaling exponent χ on the Lévy exponent β for the case of one-dimensional Lévy walks. For large β , the value of χ is close to the expected value of 1/4 while as β decreases below $\beta_c = 2$ the value of χ increases and appears to saturate at a value close to 1/3. Also shown for comparison is the rate-equation prediction (9) for χ as a function of β (dashed line). As can be seen, there is reasonably good



FIG. 3. Island-density scaling exponent χ as a function of the Lévy exponent β for the case of one-dimensional Lévy walks and flights. The symbols correspond to simulation results while the solid curve [Eq. (9)] and dashed curve [Eq. (12)] correspond to the rate-equation predictions.



FIG. 4. Pictures of clusters formed during submonolayer deposition with two-dimensional Lévy diffusion at coverage $\theta = 0.2$ with $D/F = 10^9$ (picture size L = 300). (a) $\beta = 4.0$ (b) $\beta = 0.5$.

agreement between the simulation results and the rateequation prediction. However, the simulation results tend to be consistently slightly lower (about 0.01) than the rateequation prediction for both large and small β . This may be due in part to finite-size effects as well as to the fact that we are not in the fully asymptotic (large D/F) limit.

B. One-dimensional Lévy flights

We have also carried out simulations for the case of deposition with diffusion via one-dimensional Lévy flights. As already mentioned, in this case the simulations were the same as for the walks, except that the diffusing adatoms were allowed to hop instantaneously the length of the selected hop rather than at a finite velocity.

Figure 3 shows a summary of our results for $\chi(\beta)$ for this case. As can be seen, for $\beta > 2$ there is very little difference between the results for Lévy flights and Lévy walks. However, for $\beta < 2$ the value of χ is significantly higher for Lévy flights than for Lévy walks as expected due to the large hop length. Also shown in Fig. 3 is the rate-equation prediction (12) for $\chi(\beta)$ for one-dimensional Lévy flights. As can be seen, there is very good agreement between our simulation results and the prediction of the scaling theory.

C. Two-dimensional Lévy walks

Figure 4 shows typical pictures of the clusters formed by Lévy walks in two dimensions at coverage $\theta = 0.2$ both for



FIG. 5. Island density *N* as a function of coverage for the case of submonolayer deposition with diffusion via two-dimensional Lévy walks for $D/F = 10^6$ and $\beta = 4.0$ (Brownian diffusion), $\beta = 1.33$ (enhanced diffusion), and $\beta = 0.67$ (ballistic diffusion).

 $\beta = 4.0$ corresponding to Brownian diffusion as well as for $\beta = 1/2$ corresponding to enhanced or ballistic diffusion. For large β the clusters look dendritic and similar to those obtained in previous simulations [6,8] of deposition with shortrange diffusion and irreversible attachment and without cluster relaxation. In this case, the fractal dimension d_f of such clusters is less than 2 but is close to that obtained in ordinary diffusion-limited aggregation (DLA) [24] for which $d_f \approx 1.7$ in two dimensions. However, as Fig. 4(b) shows, for $\beta < 2$ the clusters formed are significantly more compact and appear to have a fractal dimension which approaches 2 for small β . This is due to the "persistence" of the walks for $\beta < 2$, which allows particles to penetrate more deeply into a cluster than for the case of short-range diffusion. We note that the dependence of the fractal dimension of single DLA clusters grown via Lévy walks on the walk exponent β has been previously studied [25,26], and found to be in qualitative agreement with the results above.

Figure 5 shows typical results for the dependence of the island density on coverage for three different values of β for the case of two-dimensional Lévy walks. As for the case of one-dimensional walks, the overall island density decreases with decreasing β due to the increase of the diffusion length. Similarly, the coverage corresponding to the peak island density is significantly lower in two dimensions than in one dimension. Typical results for the peak island density as a function of D/F are shown in Fig. 6. As expected, the value of χ is close to 1/3 for $\beta > 2$ but increases above this value for $\beta < 2$.

Figure 7 shows a summary of our simulation results for $\chi(\beta)$ for two-dimensional Lévy walks along with the rateequation prediction, Eq. (10). As can be seen, there is reasonable agreement with Eq. (10). However, for very small β as well as for $\beta > 2$ the simulation results are slightly higher. This may be due in part to the effects of logarithmic corrections that are known to occur in two-dimensions [6,7]. The "fractal" dimension of the clusters discussed above may also play a role in the slightly increased values of χ , since it has been shown [4,10] that for submonolayer deposition with



FIG. 6. Maximum island density N_{max} as a function of D/F for deposition with diffusion via two-dimensional Lévy walks for the same values of β as in Fig. 5. Dashed lines correspond to power-law fits.

ordinary (Brownian) diffusion (d=2) and clusters with fractal dimension $d_f < 2$, the asymptotic value of χ is larger than 1/3 and is instead given by $\chi = 2/(4+d_f)$. This gives $\chi \approx 0.35$ rather than $\chi = 1/3$ in the low-coverage, large D/F, large β (Brownian diffusion) limit for which $d_f \approx 1.7$. However, for small β ($\beta < 2$) this effect should be reduced since one expects $d_f = 2$ for $\beta \le 1$.

D. Two-dimensional Lévy flights

We have also carried out simulations for the case of twodimensional Lévy flights. Figure 7 shows our simulation results for $\chi(\beta)$ for this case along with the rate-equation prediction Eq. (12). As for the one-dimensional case, for $\beta > 1$ the results for two-dimensional flights are relatively close to—although slightly higher than—those for two-



FIG. 7. Island-density scaling exponent χ as a function of the Lévy exponent β for the case of two-dimensional Lévy walks and flights. The symbols correspond to simulation results while the solid curve [Eq. (10)] and dashed curve [Eq. (12)] correspond to the rate-equation predictions.



FIG. 8. Island-size distribution scaling functions $f(s/S) = N_s S^2/\theta$ for the case of one-dimensional Lévy walks. (a) $\beta = 4.0$. (b) $\beta = 4/3$.

dimensional walks. However, for $\beta \le 1$ —for which the average hop length diverges—the exponent χ is significantly larger for Lévy flights than for Lévy walks. Over this range there appears to be reasonably good agreement between our simulation results for $\chi(\beta)$ for the case of two-dimensional Lévy flights and the rate-equation prediction (12), although the simulation values are still a little bit higher than the rate-equation prediction. This may be due in part to the existence of logarithmic corrections in two dimensions, as well as to the fractal cluster dimension $d_f < 2$ for $\beta > 1$.

E. Scaling of the island-size distribution

In addition to the scaling of the island density N, we have also studied the island-size distribution $N_s(\theta)$ [where $N_s(\theta)$ is the density per site of islands of size *s* at coverage θ] as a function of coverage and the Lévy exponent β . In particular, we have measured the scaled island-size distribution [9,10,27,28]

$$f(s/S) = N_s(\theta) S^2/\theta, \qquad (15)$$

where S is the average island size, for both one- and two-



FIG. 9. Island-size distribution scaling functions $f(s/S) = N_s S^2/\theta$ for the case of two-dimensional Lévy walks (a) $\beta = 4.0$. (b) $\beta = 4/3$.

dimensional Lévy walks and flights. Over a range of coverage in the precoalescence scaling regime, the scaled islandsize distribution is expected to be independent of both the ratio D/F and the coverage θ [7–11].

Figures 8 and 9 show typical results for the scaled islandsize distribution for the case of Lévy walks in one and two dimensions. As can be seen from Fig. 8, there is little difference between the scaled island-size distribution for the case of Brownian diffusion ($\beta = 4$) and long-range Lévy diffusion $(\beta = 4/3)$ for the case of one-dimensional Lévy walks. However, for the case of two-dimensional Lévy walks (Fig. 9), there is a small but noticeable difference between the Brownian diffusion ($\beta = 4$) case and long-range Lévy diffusion $(\beta = 4/3)$ case. In particular, for the case of long-range diffusion the peak of the scaled distribution appears to be somewhat lower, while there are relatively more small-size clusters. This is most likely due to the increased compactness of two-dimensional clusters for small $\beta < 2$, which leads to decreased island-island coalescence in the case of long-range diffusion. Similar results for the scaled-island-size distribution have been obtained for the case of one-dimensional and two-dimensional Lévy flights.

VI. CONCLUSIONS

We have presented the results of kinetic Monte Carlo simulations for the scaling of the island-density and island-size distribution in the case of submonolayer deposition with long-range Lévy diffusion in one and two dimensions. Results were presented for both Lévy walks and Lévy flights and were restricted to the case of a critical island size of 1 corresponding to stable clusters of size 2 or greater. Using scaling arguments and a rate-equation analysis, an analytical prediction for the dependence of the island-density scaling exponent χ on the diffusion exponent β was also derived and compared with our simulation results.

For the case of submonolayer deposition with adatom diffusion with a Lévy exponent $\beta > 2$, we found good agreement in both one and two dimensions between the standard (short-range hopping) rate-equation predictions for the scaling exponent χ relating the dependence of the island density on the ratio D/F of the diffusion rate to the deposition rate. However, for the case of submonolayer deposition with longrange Lévy diffusion ($\beta < 2$) the scaling exponent $\chi(\beta)$ was found to be significantly larger than for the case of Brownian diffusion and to increase with decreasing β . Reasonable agreement was found between our simulations and our rateequation predictions for $\chi(\beta)$ for the case of Lévy walks in both one and two dimensions. In particular, while our simulation results for χ for $\beta > 2$ were close to the standard values of 1/4 (1/3) in one (two) dimensions, for $\beta < 2$ the value of χ was found to increase with decreasing β and to approach our rate-equation prediction of 1/3 (2/5) for small β in one (two) dimensions. We also found good agreement with our rate-equation predictions for $\chi(\beta)$ for Lévy flights in one and two dimensions, for which χ was somewhat higher than for walks.

In contrast, the scaled island-size distribution was found to be almost the same for the case of long-range diffusion as for short-range diffusion. This indicates that the main effect of long-range diffusion is to increase the scaling exponent χ rather than to alter the island-size distribution. Consequently, in experiments (such as in liquid-phase epitaxy and/or electrochemical deposition) in which long-range diffusion may play a role, the principal observable effect may be the increased sensitivity of the island density to the deposition rate due to the increased value of χ .

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