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**A kinetic mechanism for the transformation of
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by

Z. Zhang, Y.-T. Lu and H. Metiu

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A kinetic mechanism for the
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on a vicinal Si(100) surface.

Zhenyu Zhang, Yan-Ten Lu, and Horia Metiu

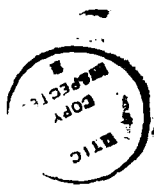
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Abstract: We perform energy calculations with the Stillinger Weber potential to determine (a) how the Si atoms deposited on a Si(100) surface with single layer steps migrate to produce a surface with double layer steps and (b) why the system does not evolve by the usual step flow mechanism.



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The properties of stepped Si(100)-2x1 surfaces have been studied extensively^[1-15]. There is experimental evidence^[6,8-9] that single atom layer steps (S steps) can be transformed into double atom layer steps (D steps) by changing the temperature or by depositing Si on the surface. We are particularly interested here in the experiments of Hoeven, Lenssinck, Dijkkamp, van Loenen and Dieleman^[6]. They started with a Si(100)-2x1 surface having S steps. Such a surface has two kinds of terraces, denoted here TA and TB (Fig. 1). They can be distinguished by the orientation of the dimer rows: the rows on the TB terraces are perpendicular to the steps and those on the TA terraces are parallel (Fig. 1). The steps between terraces are also of two types: the SA (SB) step^[2] has an TA (TB) terrace above it (Fig. 1). Hoeven *et al.*^[6] deposited half a monolayer of silicon on this surface and examined the result by STM. They found that the surface has D steps with the dimer rows perpendicular to the steps (TB terraces). The width of these terraces is twice the width of those on the starting surface.

The formation of the D steps after the deposition require a complicated atomic motion: the atoms which have landed on a TA terrace must stay there, while the ones that have landed on a TB terrace must move onto a neighboring TA terrace (Fig. 2). In this article we present energy calculations which support this mechanism and provide details of the movement of the atoms across steps, from one terrace to another. We use the Stillinger Weber potential to determine the binding sites of the adsorbed Si atoms at the step and at the terrace edge, and the energy barriers for the site to site jumps. The latter quantities are used to estimate the site to site jumping rate constants. To

map out the most likely growth pathway we assume that the atoms will tend to execute most often the jumps having the highest rates. In this way we can conjecture the main features of the growth mechanism without having to perform detailed and time consuming kinetic simulations^[17].

The binding energy calculations are performed by allowing the adsorbed atom, and all the lattice atoms that can influence the result, to relax to their minimum energy positions. We check to ensure that the finite size of the sample does not affect the results. The energy barriers separating two binding sites are calculated by making a sequence of small displacements that take the adsorbed atom from one binding site to another. For example if the initial site is the one marked C in Fig. 1 and the final site is the one denoted D_1 , then we displace the atom from C along the straight line leading to D_1 . Let us denote the displaced atom's coordinate along this direction by x and the other coordinates by y and z . After each small increase of x , we keep x fixed and vary the coordinates of all the other surface atoms, as well as the coordinates y and z of the displaced atom, until they reach the positions corresponding to the minimum energy. This procedure is repeated until the atom reaches the final destination D_1 . This gives us the minimum energy pathway for a jump from the initial to the final site, and the energy barrier hindering the jump. Often two sites can be connected by several pathways and care must be taken to find the one with the lowest energy^[14].

The adsorption sites, corresponding to local energy minima for an adsorbed atom, are shown in Fig. 1 and are denoted by A, B, C, D, E and F^[18]. Subscripts are used to label identical sites located in different surface unit cells (e.g. F_1 , F_2 and F_3 are identical by symmetry). The atoms arriving from the

vacuum have a propensity to stick at the sites B, C or D^[18]. They can move rapidly along a dimer row by executing the jumps $B \rightarrow E \rightarrow C \rightarrow E \rightarrow B$ with side trips to D^[18]. Jumps in the direction perpendicular to the dimer row occur only very rarely^[18].

With these in mind we examine the fate of the atoms that have landed on a TA terrace (i.e. located above an SA step). According to the model proposed in Fig. 2 we expect the transformation from single to double step to take place if the deposited atoms can leave the TA terrace only with great difficulty. We examine below the energy barriers along several pathways on which an atom might leave the TA terrace and we find them all (see below) to be rather large.

If we start an atom from the site B, shown in the left upper corner of Fig. 1, we must provide a large amount of energy to push it towards the site A located near it. This happens because the atom at B binds to the atoms 1 and 2 of the substrate to form a trimer. When pushed towards the site A, an atom located at B drags the 1 - 2 pair with it. When the energy required for performing this displacement reaches 1.81 eV the bond between the adsorbed atom and the 1-2 pair breaks, the pair returns to its original position in the surface dimer row, and the adatom moves into the A site. This large energy barrier prevents the B to A displacement from occurring frequently enough to be an important factor in adsorbate mobility. The energy barrier to move from the site A to the site F₁ located nearby is only 0.30 eV.

An adsorbed atom starting in C (Fig. 1) must cross a 0.15 eV barrier to reach the nearby D₁ site, and then a barrier of 0.79 eV to reach F₂.

An atom that starts from E and is being pushed towards the neighboring

site D_2 , will drag the 3 - 4 pair along until the bond between the adsorbed atom and the pair is broken; then the pair returns to its initial position and the atom reaches D_2 . The energy barrier for this move is 1.70 eV. A further move from D_2 to F_3 has a barrier of 0.79 eV.

All three displacements examined above bring the adsorbed atom to an F site. The barriers for some of these displacements are quite high. The fastest route by which an atom located on a dimer row reaches an F site is to move on a D site (the barriers for $B \rightarrow E \rightarrow C \rightarrow D$ are low) and jump from there to the neighboring F site, with a barrier of only 0.79 eV. Other pathways, such as a the jump $C \rightarrow D$ towards the step, encounter a larger barrier and are therefore infrequent. However, an atom that has reached an F site has difficulty moving towards the edge of the TA terrace (i.e. in a direction perpendicular to the dimer rows). The most efficient pathway for such a move is a displacement parallel to the step, to a place half way between two F sites, followed by a displacement towards the step to reach a C site. The energy barrier for such a move is 1.25 eV. All other moves towards the terrace edge have barriers of approximately 3 eV.

Thus, the fastest pathway for moving across the TA terrace encounters a barrier of 1.25 eV. Compared to other relevant barriers (see what follows) in the system, this is rather high. Therefore, we conclude that most of the atoms landing on the TA terrace will be stuck there.

We examine next the behavior of the atoms landing on the TB terraces. The model proposed above (see Fig. 2) suggests that the transformation from single to double steps can take place if these atoms can leave the TB terrace to descend to a TA terrace (across the SB step) and/or to climb up to the neighboring TA terrace (across the SA step). We show below that both these events take place frequently.

Previous work^[18] has shown that an adatom landing on a Si(100) surface is most likely to stick to a site located on top of a dimer row (i.e. the sites B, C or E). The largest barrier against the motion along the dimer row is 0.29 eV. As a result

the adsorbed atom moves quickly along the dimer row. Since on a TB terrace the dimer rows are perpendicular to the steps, the adsorbed atom has no difficulty reaching the step. Therefore, to decide whether the atoms leave the TB terrace we must find whether they will cross the step onto a neighboring terrace. We consider first an atom located on the C_1 site near the SB step on the TB terrace (see Fig. 1). The largest barrier encountered by an atom moving from C_1 to B_1 is 0.44 eV. The B_1 site differs from the other B sites on the dimer row (such as B_2 in Fig. 1) because it is located at the edge of the terrace. For this reason the 0.44 eV barrier to reach B_1 from C_1 differs from the 0.29 eV barrier encountered by an atom moving from C to the B_2 site (see Fig. 1). Thus, the atom approaching the SB step on the TB terrace prefers to return to the terrace rather than occupy the B_1 site. However, at a cut angle of 0.52 degrees the TB terrace has approximately 38 sites. At 750 K the C_1 site will be visited 10^9 times per second. Because of this the chance that the atom reach the site B_1 is fairly high.

To move down the SB step from B_1 and reach the site E_1 [Fig. 1(b)] the adatom must overcome two barriers, one of 0.40 eV and the other of 0.84 eV. The binding energy to the E_1 site is large and the barriers for moving back across the SB step on the TB terrace or away from E_1 on the TA terrace, are of about 2 eV. This result is consistent with the experimental observation of Mo and Lagally^[13], who found that the SB step traps the adsorbed particles very efficiently.

The atom bound at E_1 can easily move along the SB step. The largest barrier encountered along the way is 0.37 eV. This is expected since the pathway along the step is on top of a dimer row, where the mobility is rather high. The presence of the step nearby does not changes this mobility by much: the largest barrier on the string near the step is 0.37 eV, while the largest barrier when moving on a string that is not near the SB step is 0.29 eV. Thus, an atom descending from a TB terrace across the SB step to the E_1 site will move along the

step until it encounters another adatom on the same string and form a dimer. This dimer will line up with the dimer row of the TB terrace, leading thus to step flow and terrace growth. The atom at an E_1 site can also interact with an atom located at a site of type B_1 , at the edge of the TB terrace [see Fig. 3(a)]. The presence of an adatom at an E_1 site (close to the SB step) helps an atom at the B_1 site nearby (on the upper TB terrace) to jump down across the step. If we push the atom 1 located at a B_1 site towards the SB step it will form a dimer [Fig. 3(b)] with the atom 2 located at E_1 . The presence of atom 2 lowers the barriers encountered by 1. Unfortunately the dimer formed by these two atoms is in the "wrong position". This error can be corrected by rotating the dimer to the position shown in Fig. 3(c). This move requires overcoming a 0.73 eV barrier.

In summary, according to these calculations the atoms landing on a TB terrace can step down, across the SB step bordering it, to the TA terrace below. Upon reaching this terrace these atoms can easily move along the step until they encounter another atom on the same row and form a dimer with it. Another option is to assist an atom to descend from the TB terrace, and bind to it to form a dimer. In either case the dimer will extend the TB terrace. This behavior is consistent with the mechanism outlined in Fig. 2.

Now we examine what happens when an adatom moves on a TB terrace and approaches an SA step. The mechanism proposed in Fig. 2 assumed that such atoms can cross the SA step to climb on the TA terrace above it.

Since the dimer rows on a TB terrace are perpendicular to the SA step the adatoms located on this terrace have no difficulty reaching the step, by moving rapidly on top of the dimer rows. Such an atom, denoted 1 in Fig. 4(a), reaches the position G of Fig. 4(a) without difficulty, by moving on top of a dimer row as indicated by the arrow (Fig. 4a). Once there, the atom is prevented by a barrier of over 2 eV from moving back onto the TB terrace (away from the SA step). The barrier for moving along the SA step to the S site (see Fig. 4) is 0.36 eV. Once there the atom is trapped. Before hopping to S it can also participate in what we called

previously^[14] an exchange process. The outcome of this process is shown in Fig. 4(b): the atom 1 has replaced the atom 2, and the latter has been lifted up on the TA terrace. The outcome is that an atom left the terrace TB and one appeared on the terrace TA above. This exchange process must overcome an energy barrier of 1.07 eV.

The exchange process is aided if another adatom, N, approaches [Fig. 4(c)] the adatom 1, located at the G_1 . The energy barriers are such that N has no difficulty in getting close to 1 and forming the dimer shown in Fig. 4(d). If we push the atom N towards the step the atom 1 will engage in an exchange process whose outcome is shown in Fig. 4(e). The presence of N lowers the barrier for the exchange process from 1.07 eV to 0.51 eV.

The atom placed by the exchange process on top of the dimer row (on the TA terrace) can easily move in a direction parallel to the step, leaving behind the atom N in the configuration shown in Fig. 4(a).

We note that the dimer 1-N shown in Fig. 4(d) is not very stable: the N atom can easily move away onto the TB terrace or the 1 atom can use the exchange process to break away from N. Therefore, the SA step is less effective than the SB step in trapping the adatoms reaching it from the lower terrace. This is consistent with the observation of Mo and Lagally^[13] that more islands are formed on a TA terrace near the SA step (left hand side of Fig. 4) than on the TB terrace near the SA step (right hand side of Fig. 4).

The calculations presented here were performed to help us understand the kinetic factors controlling the transformation of S steps to D steps, upon deposition of half a monolayer of silicon. We have found several kinetic features that control the outcome of this growth process. The first is the anisotropy of the adsorbate motion on the Si(100)-2x1 surface. An adatom located on top of a dimer row moves frequently along the row, and rarely in the direction perpendicular to it. This affects strongly the particle transport on the terraces. An atom adsorbed on a TB terrace, on which the dimer rows are perpendicular to the step, have no trouble

reaching the step or the terrace edge. On a TA terrace the dimer rows run along the terrace and the transport of the adsorbed particles to the edge or the step is extremely slow. This effect alone will make it difficult for the adsorbed atoms to leave the TA terraces. We note that the possibility that the migration of Si on the Si(100)-2x1 surface is anisotropic has been recognized by many authors, and recent experimental evidence for this has been provided by Lagally's group^[9]. The anisotropy suggested by the present calculations seems to be more pronounced than what was previously believed.

The anisotropy of diffusion is only half the story. It explains the tendency of the adsorbed particle to stay on the TA terraces. To understand the formation of the double layer steps we must also find why and how the particles landing on the TB terraces leave them and move to the TA ones. Our energy calculations indicate that a particle can leave a TB terrace by climbing down (across the SB step) onto the neighboring TA terrace; the largest barrier along the way is 0.84 eV. Furthermore, the atoms adsorbed on the TB terraces can reach the SA step and engage in what we call the exchange process; as a result an adatom disappears from the TB terrace and one appears on the TA one. The barrier for the exchange process is 1.07 eV if unassisted by a second atom and 0.54 eV if a second atom is nearby.

While some of the barriers quoted here are fairly high one must keep in mind that the Si deposition rate is very small (about 0.001 monolayer/sec) compared to the rate of the site to site jumps. For this reason, if the temperature is sufficiently high the adsorbed atoms have plenty of time to overcome such barriers without being bothered by other atoms which can tie them up by forming dimers, trimers and ultimately islands. If the temperature is too low, such islands are formed on terraces and prevent the formation of the double steps.

The accuracy of the Stillinger Weber potential is still uncertain^[19]. The tests performed so far^[19] indicate that it works reasonably well for the (100) surface and the liquid, while it is unable to reproduce the intricate reconstruction

of the (111) surface and the structures of some of the smaller clusters. Most of the conclusions reached by the present work are qualitative and depend on the ability of the potential to distinguish which barriers are very small and which are not.

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FIGURE CAPTIONS.

Fig.1: The positions of the atoms in the outermost layer of a Si(100)-2x1 surface having steps with a single atom height. The darker circles represent atoms closer to the reader. The dimer rows on the TA terraces are parallel to the step, and those on the TB terraces are perpendicular. An SA (or SB) step has an TA (or TB) terrace above it. The small black dots indicate adsorption sites for a Si atom. The sites labelled by the same letter but different subscripts are identical by symmetry. The long arrows show the direction of rapid migration.

Fig.2: A schematic representation of a mechanism for the transformation of single atom high steps into two atom high steps with B type terraces. (a) Shows the starting stepped surface on which one deposits half a monolayer of Si. (b) Shows the final surface having two atom high steps with B terraces. (c) Shows a possible kinetic mechanism thorough which the transformation from (a) to (b) takes place.

Fig. 3. A mechanism for stepping down across an SB step. (a) An atom 1 approaching the SB step by moving along a dimer row on the TB terrace will be aided by the atom 2 located on a site E_1 near the step on the TA terrace. (b) The dimer formed by the atoms 1 and 2. The dimer has the "wrong" orientation. (c) The position reached by the dimer after rotation. This dimer extends now the SB terrace. The black and stripped atoms are in the same layer.

Fig. 4. The exchange mechanism by which an atom starting on the TB terrace near the SA step is transferred onto the SA terrace and the manner in which a second atom can help this transfer. (a) The atom 1 has approached the step by moving along the dimer row on the TB terrace as indicated by the arrow. (b) The

positions of the atoms 1 and 2 after the exchange took place. (c) The atom 1 at the site G can be helped to perform an exchange process by the atom N approaching it along the dimer row. (d) The position of the atoms 2, 1 and N before the exchange. (e) The position of the atoms 2, 1 and N after the exchange. The atom 2 is on the TA terrace. (f) After the exchange took place the atoms 2 and N can easily move as indicated by the arrows.

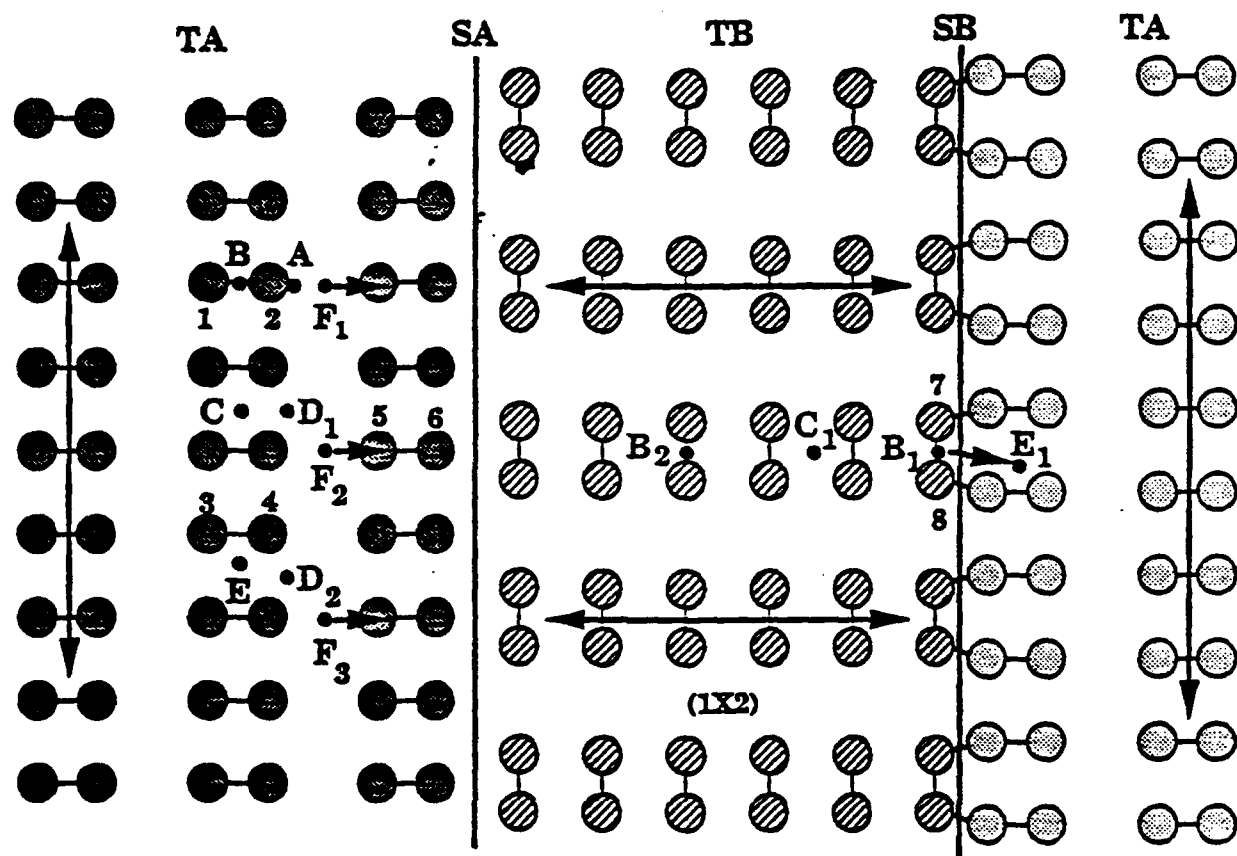


Fig. 1

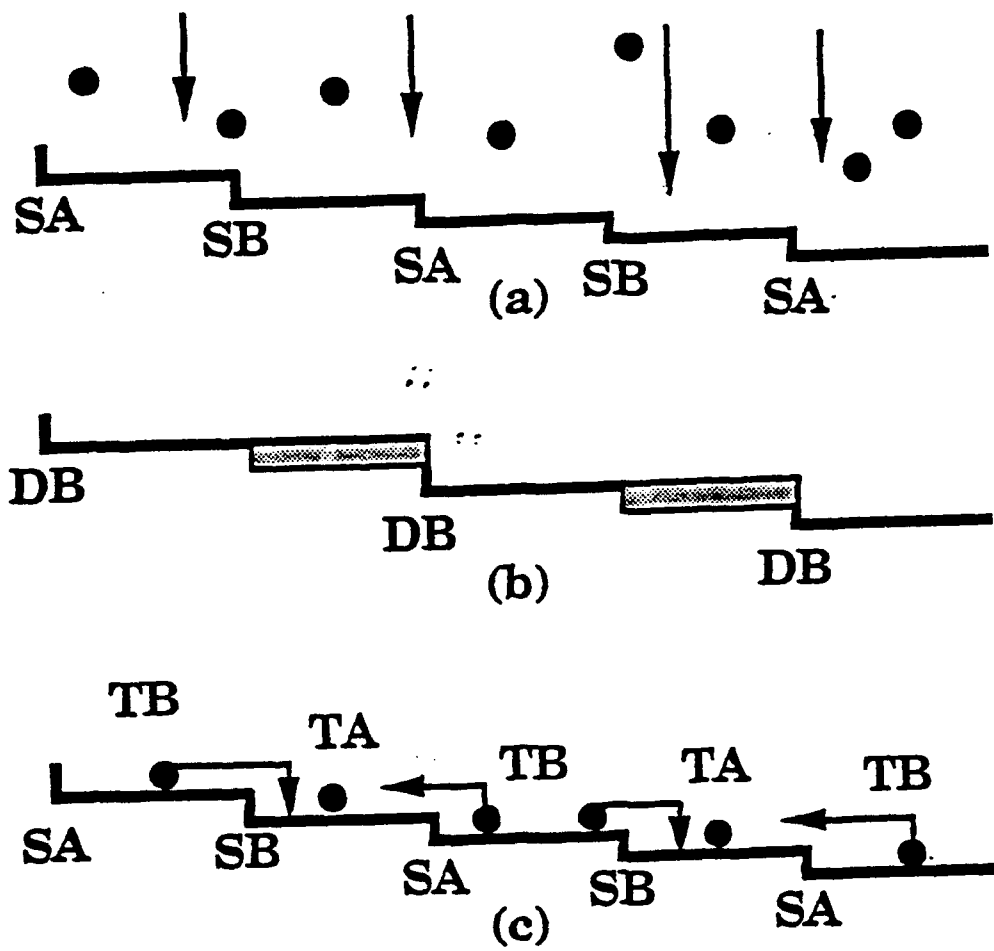


Fig. 2

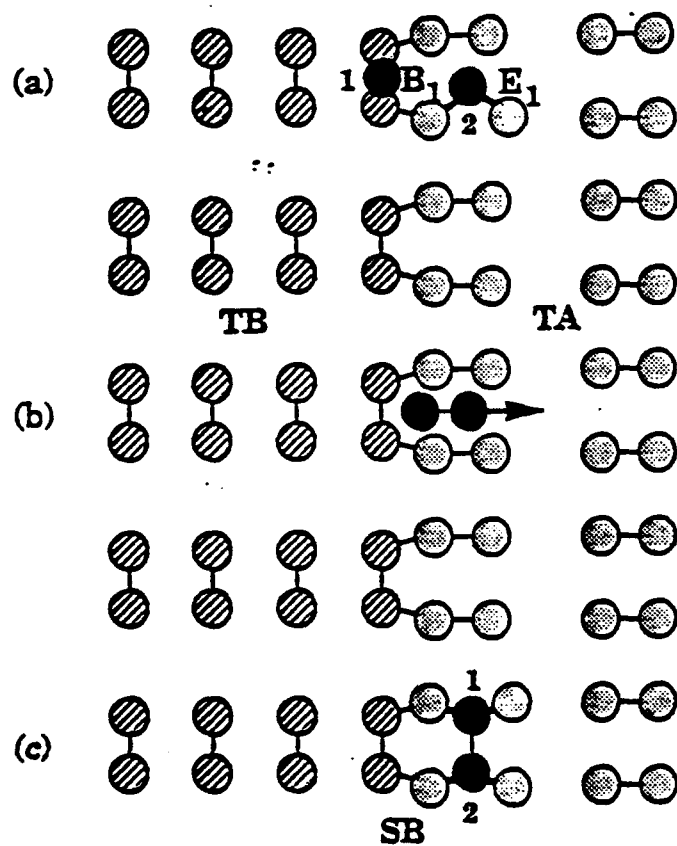


Fig. 3

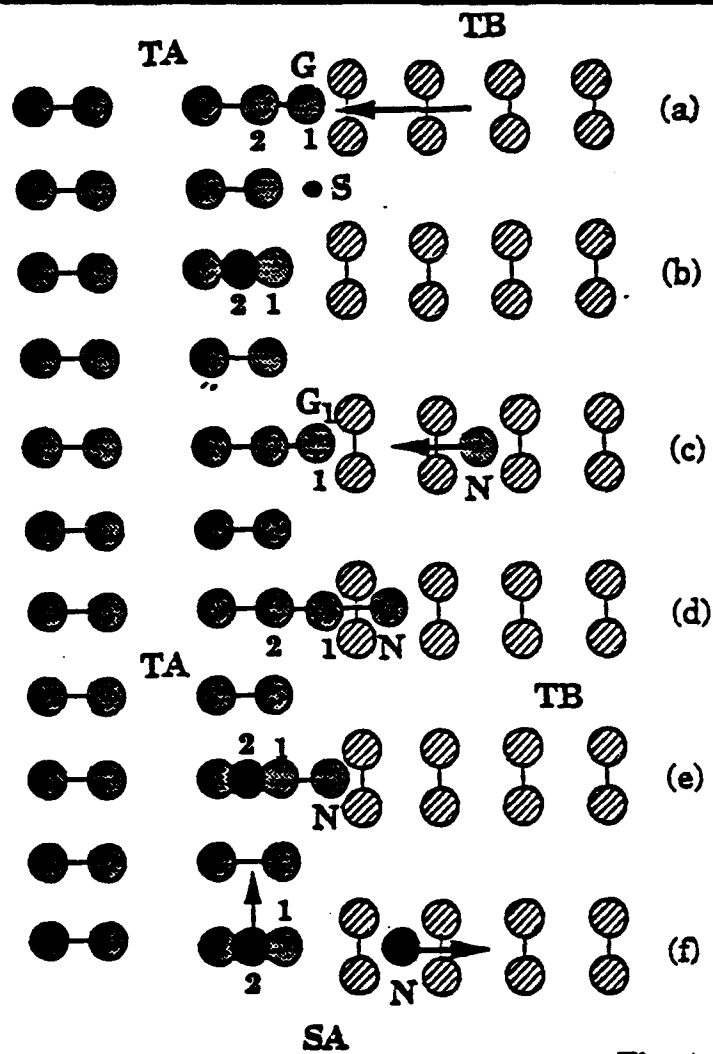


Fig. 4

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