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ARTICLE *in* ISRAEL JOURNAL OF CHEMISTRY (ONLINE) · JANUARY 1990

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Dynamics and Potential Surfaces of Small Clusters

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(Received 26 April 1989)

Abstract. Some of the tantalizing puzzles regarding clusters are described. Some specific results concerning the dynamical behavior of clusters of particular sizes and having various potentials are described, to give some insight into the relationship between the form of a potential and the kind of behavior it yields. Solid-liquid, solid-soft solid, surface melting, and a remarkable single "subsurface layer mobility" all can be found in particular instances.

INTRODUCTION

The bridge between molecular matter and bulk matter is being built now by the lively community of chemists and physicists working on those things we call clusters. Neither so large that we can call them infinite, boundless collections of particles, nor so structurally and stoichiometrically constrained that we can call them molecules, their tractability has given us many new insights into the behavior of bulk matter. At the same time, clusters offer possibilities for making new materials and devices whose properties are engineered at the molecular level. Both Joshua Jortner and Raphael Levine have made important contributions to our understanding of the dynamical behavior of these small systems, particularly of the kinds of states small systems may display when energized. This essay is devoted to some aspects of that issue: how do the forms of the interaction potential between particles and the potential energy surface, and the manner in which we observe or probe the behavior of a cluster, affect the properties we see and attribute to the cluster?

Theoretical investigations have shown that clusters may have solid-like and liquid-like forms, that the solid-like forms may be rigid or soft and floppy, and that different forms, such as homogeneous, "single-phase" solid-like and liquid-like clusters, may coexist in dynamic equilibrium like chemical isomers over a range of temperatures at constant pressure.¹ Simulations have borne out the inferences, in fact have in some cases preceded and stimulated them. Some of these results, such as the coexistence of well-defined solid and liquid forms, have heretofore only been found

in clusters of selected sizes and with specific interaction potentials, notably the Lennard-Jones in this case.^{1,2} Others, such as coexisting rigid and floppy solid forms, have been found for clusters of single kinds of atoms with several different potentials and of alkali halides composed of ions interacting via Born-Mayer potentials.^{3,4} Enough specific cases have been studied now to induce us to ask how sensitive such behavior is toward the choice of potential, through either its analytic form or the values of its parameters. There are not yet any rules about the generality of different kinds of coexistence or of intermediate, slush-like behavior.

Some puzzles have emerged, such as an NaCl cluster which seems to be solid on one side and liquid on the other, rather than solid on the inside and liquid on the outside.⁴ This implies a substance whose melt does not wet its own solid, an uncommon and striking phenomenon. A cluster exhibiting this behavior could show true superheating of the solid, the counterpart of the well-known supercooling of the liquid. The normal situation of a less dense liquid which wets a more dense solid of the same substance has recently been described by Reiss et al.⁵ There does seem to be a direct relation between the stability of solid-like and liquid-like forms of clusters and the metastability of bulk, homogeneous matter, either superheated or supercooled.⁶

Other puzzles concerning clusters themselves and their properties have emerged from the study of their phase changes. Some of the most tantalizing have been the questions of what forms, notably monotonic or S-

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shaped, may be taken by the caloric curves of $\langle T(E) \rangle$ and $\langle E(T) \rangle$,⁷ and what are necessary and sufficient conditions for these two caloric curves to be superimposable or to be different.

Still other puzzles revealed or answered by the study of clusters bear on the properties of bulk matter. One is the nature of electrical and magnetic properties of clusters, with such manifestations as the transition from insulating behavior in small clusters to metallic, conductive behavior in larger clusters.⁸ Why and how this transition occurs remains unexplained. Closely related is the question of chemical reactivity of atoms in clusters, and of how their reactivity depends on the size and phase or geometry of the cluster.⁹ Again the issue of phase changes appears: in order to interpret the kinetics of reactions, we will assuredly have to address the kinetics, as well as the thermodynamics, of phase changes and geometric transformations in clusters.

All these are some of the tantalizing issues that have attracted many of us to study clusters. In the following discussion, we shall present some results that bear on some of these issues and relate them to the previous findings of others. In particular, we shall discuss here some comparisons showing how dynamical effects are affected by the choice of interaction potential, and how solid-solid and solid-liquid phase changes appear in some kinds of clusters. We address in particular some aspects of the sensitivity of potential surfaces and dynamics to the choice of potential.

EFFECTS OF THE POTENTIAL

The classic work on how potentials determine the stable geometries of clusters was the series of studies by Hoare and Pal.¹⁰ Their work and related studies concentrate on central, pairwise potentials suitable for rare gas clusters and, in Born-Mayer-Rittner form, for alkali halides. The first object of such studies was the determination of the minima, both their geometries and their energies. More recently the focus has moved to studying dynamics and, still in its infancy, the connection between dynamics and the form of the surface. We shall emphasize some of these latest two aspects, but a few comments are in order regarding structural questions.

While central, pairwise potentials have been used to represent atomic clusters of metallic, semiconducting, and insulating elements, the reliability of results of such calculations is still quite unclear. That more elaborate potentials are needed for accurate predictions of spectra goes without saying; central, pairwise potentials cannot reproduce the distortions of a Jahn-Teller effect, for example. However, it will be important to

learn how well central, pairwise potentials can reproduce the main features of multidimensional potential surfaces for purposes of molecular dynamics and Monte Carlo simulations. Full quantum-mechanical simulations are possible for such systems, in principle. However, it is simply unrealistic now to consider paying the cost of systematic simulations by molecular dynamics in which the forces on the nuclei are computed at each step from quantum mechanics of the entire electronic structure. A few *tours de force* will surely be done this way,¹¹ but the best use at present for such approaches is to guide the parametrization and choice of form of simpler, more efficient, and approximate potentials.

The search for all the minima of a potential surface was long considered important. For small clusters, this is very likely so; for large clusters, it is certainly not important. Where is the dividing line? Why do we need to know about all the minima on a surface? Better, we should ask "What information do we need in order to infer a particular property from knowledge of the potential surface?" We can begin to answer this by looking at some relatively small clusters and seeing what minima they exhibit. Finding minima was based, for a long time, on systematic exploration of growth sequences, starting with a stable, small core and adding atoms one at a time, determining the site of lowest potential energy to which each could be added. This procedure, we have found, is only adequate for finding a lower bound on the number of minima. To assure that one finds all the *important* minima, not even all the minima at or below a specified energy, it is important to supplement the use of growth sequences with a combination of molecular dynamics (to allow the molecule to search its configuration space) and quenches, coupled with steepest-descents (to bring the phase point to the potential surface and, from there, down to the minimum of the catchment basin in which the point happens to lie when the quench is carried out).

Hoare¹⁰ reviewed the number and kinds of minima found for simulated argon clusters up to Ar₁₃, at least those known in 1979. There are four geometrically different kinds of minima for Ar₇ based on a Lennard-Jones potential and only three are reported for a Morse potential. We have found an additional minimum for the Morse potential. Likewise, we have found a total of nine minima for Ar₈ represented by a Morse potential, compared with only five reported by Hoare.¹⁰ For Ar₁₃ represented by a Lennard-Jones potential, Hoare and Pal originally found the correct ground state but missed the defect structures corresponding to the first three potential minima above the lowest. These were found

in the more extensive search of Hoare and McInnes.^{10,12} They appear naturally and unambiguously from molecular dynamics searches¹³ because they are the minima of the first catchment basins visited when an Ar₁₃ cluster is given enough energy to escape from its icosahedral lowest energy well. In larger clusters, it is possible, perhaps even likely, that some important, low-energy structures will be found which do not arise from systematic growth sequences. Three examples may have been found already of what appear to be *lowest energy* geometries that have been revealed¹³ by molecular dynamics but not by growth sequence searches. These are the most stable forms of Ar₁₇, Ar₂₂, and Ar₃₃.

The number of geometrically different stable forms of a cluster rises rapidly with N , the number of particles in the cluster. Hoare¹⁰ quotes 2, 4, 8, 18, 57, 145, 366, and 988 as the numbers of such structures for N from 6 through 13. But to understand dynamics on a potential surface, we must be concerned with passages from well to well, including those that only permute atoms among equivalent sites. This means that we should be concerned with the number of geometrically identical, permutationally different potential wells, in addition to the number of geometrically different wells. Looking at these numbers makes us realize quickly how important it is to try to use summarizing indices and properties beyond the detailed atomic level to describe clusters. For the Ar₇ cluster with Lennard-Jones potentials, there are 504 permutational isomers of the pentagonal bipyramid, the structure of lowest energy. There are 1680 permutational isomers of the monocapped octahedron, the same number for the tricapped tetrahedron (or tetratetrahedron), and 5040 such isomers of the highest energy structure, the “skew” or tetrahedron + two caps + one edge. The total is 8904 isomers for Ar₇, surely a small cluster if one wants to think of deriving bulk properties from clusters. The numbers of geometric and permutational isomers have been conjectured to increase exponentially with N . We can only infer from this that we need conceptual tools different from and simpler than complete knowledge of the potential surface if we are to be able to use information about that surface.

DYNAMICS AND POTENTIAL SURFACES

One of the challenges mentioned above is establishing the connection between the form of a potential surface and the kind of dynamics a system exhibits at any specified energy or temperature. The beginnings of such understanding are emerging now in the context of clusters, probably because they, in contrast to conventional molecules, can explore large regions of the topog-

raphy of their potential surfaces without dissociating. One example is the six-particle cluster. The dynamics of this cluster has been studied by Sawada and Sugano¹⁴ with a Gupta two-body potential intended to simulate a cluster of transition metal atoms, by Blaisten-Barojas and Levesque¹⁵ with a Stillinger–Weber potential including one-, two-, and three-body terms to simulate silicon, and by ourselves with Lennard-Jones, Morse, and “embedded-atom” potentials, the first to represent rare gas clusters, the third to represent Cu clusters, and the second for general use.

With any of the potentials used thus far, the six-atom cluster shows only two geometrically distinct minima. For all the potentials but the Stillinger–Weber one, the lowest is an octahedron with an energy of -12.712ϵ (ϵ is the Lennard-Jones well depth). The other is a bicapped tetrahedron or distorted octahedron, flattened, with one edge opened, whose energy is -12.303ϵ . We find saddles between the octahedra and their associated distorted octahedra at -12.079ϵ . The Stillinger–Weber potential gives a trigonal prism for the lower energy geometry and a distorted form of this structure for the higher energy form.

Sawada and Sugano reported that at a low energy, $0.72 \times E_{\text{bulk}}$, this cluster undergoes a transition from its low-energy, solid-like form to a dynamic equilibrium between this structure and the higher-energy structure, a condition they call the “fluctuating state.” Presumably this corresponds to reaching the saddle of the Lennard-Jones system at -12.079ϵ . This is clearly not a liquid-like form because there are no permutations of the atoms among different sites. The energy of 0.72 is approximately that of the saddle separating the lowest energy structure from its own distorted version, with atoms in the same relative positions. At a slightly higher total energy, $0.70 \times E_{\text{bulk}}$, the atoms are able to pass over a higher saddle and permute themselves among different sets of octahedral potentials, each with its own collection of distorted-octahedral potentials around it. For the Lennard-Jones system, this corresponds to a saddle at -11.630ϵ .

We can think of the entire potential surface of the six-atom octahedral cluster this way. There are $6!$ assignments of six atoms to the octahedral sites, but of these, any one is equivalent by a rigid rotation to 23 others; the number of permutationally different octahedra is $6!/24$ or 30. Hence there are 30 distinct octahedral minima on the potential surface. Each can be distorted to give the higher energy structure in 12 ways, so each octahedral minimum is connected to 12 higher minima. Let us call this set of 13 wells a “clock.” There is no evidence for a saddle between any pair of minima

around the same octahedral minimum, i.e., between distorted octahedra in the same clock. However, each of the 12 minima is linked by a saddle to 4 minima of the same geometry but "attached" to a different clock. Each of these links is then one of the 720 between one clock and another. From a distorted octahedron in clock #1, one can go to clock #2 and find, from each point of #2 reachable from #1, a single other return path visiting a third clock only. One can, with this kind of examination, determine the entire topology of the energy surface at each energy.

The seven-particle cluster has been studied more extensively than the six-particle cluster, partly because of the richness of its surface with four kinds of minima, partly because it is the smallest cluster that shows clear coexistence between solid and liquid forms. Sawada and Sugano¹⁴ have studied this, as have Amar and Berry¹⁶ and Beck and Berry.¹⁷ The saddles connecting the minima, at least those reachable at energies below the energy at which vaporization is significant, have been found, and their energies and geometries have been identified.¹⁸ There are six kinds of such saddles; the lowest connects the pentagonal bipyramid (lowest energy) with the monocapped octahedron, and the next two, almost identical in energy, connect that next lowest well with the two higher energy wells, in sequence. All the saddle energies are higher than any of the minima for this species, which is relevant to its sharp solid-liquid transition. The rates of isomerization of simulated Ar₇ rise rapidly with energy: five-fold with an increase of total energy of 1%, and, in absolute terms, from 0.2 ns⁻¹ at -14.45 (in units of the Lennard-Jones well depth) to 63.7 ns⁻¹ at a total energy of -13.06, a factor of 300 with a 10% increase in energy. Such increases are, of course, like the increases in rates of chemical reactions with temperature.

We get more insight into the dynamics of clusters by looking at the distributions of residence times of the clusters in each well.¹⁶ At the lowest energies, the seven-particle cluster spends almost all the time in the lowest well; in fact up to energies at which its isomerization rate is about 8.4 ns⁻¹, which is near the top of the coexistence range, the residence times in the three highest wells are mostly well below 100 ps, while the residence times in the lowest well are roughly evenly distributed out to about 1 ns. This means that from low energies through the range of coexistence, nearly every isomerization carries a cluster away from any of the higher wells, but clusters may spend of order a hundred times the mean isomerization time in the lowest well. This is precisely what is expected if the liquid can be characterized by interwell passage at a rate approaching typical vibrational frequencies, while solids spend times much longer than that in one well. At energies corresponding to liquid-like behavior, the seven-particle cluster exhibits roughly the same distribution of residence times in the upper three wells, but the times spent in the lowest well are reduced almost an order of magnitude, being longer than the times in the upper wells by only a factor of about 10.

The 13-atom cluster has been studied in great detail and will not be discussed here,^{1,10} except to mention a bit concerning the three lowest excited wells above the icosahedral lowest well. Figure 1 shows the icosahedron and the first three excited geometries. The energies of these three are very close, whatever pure two-body potential is used. They all correspond to a single atom being removed from the 12-atom icosahedron, lifted out, and put into the center of a triangular face. There are three geometrically distinct kinds of triangular faces on an icosahedron with one vertex removed, and placement on each of these corresponds to the three kinds of "first-excited" wells. The order of energies of these three

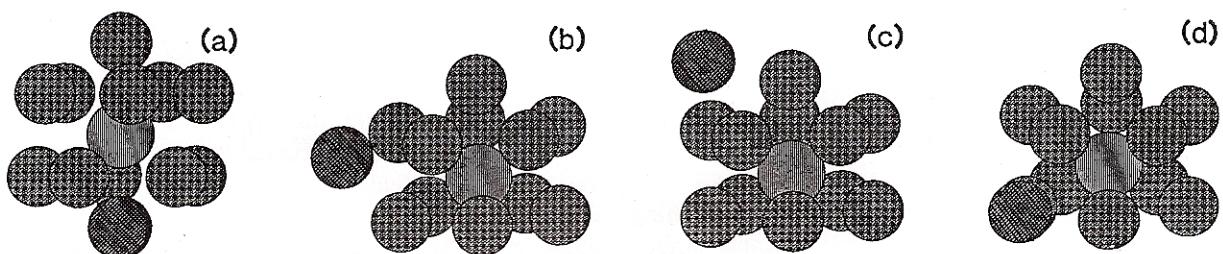


Fig. 1. The geometries of the lowest energy and next three higher energy forms of the 13-particle cluster described by a Lennard-Jones, Morse, or "embedded-atom" potential: the icosahedron (a); the first excited structure for Cu₁₃ represented by an embedded atom potential (b); the first excited structure for Ar₁₃ represented by Lennard-Jones potentials (c); the other of the three almost-degenerate first excited geometries (d). The central atom and the promoted atom are shown shaded differently from the rest for purposes of indicating the geometries; all the particles are intended to be identical.

does depend on the potential; for a Lennard-Jones potential, the lowest of the three has the displaced atom on one of the faces furthest from the vacancy, while the lowest of the three for Cu₁₃ simulated by an embedded atom potential¹⁹ has the promoted atom on a face with one vertex at an atom adjacent to the vacancy. The mechanism for production of this set of levels is not necessarily the most obvious. For the Lennard-Jones case, one atom is extruded from the shell of 12 by a collective squeezing effect by the other 11, a sort of atomic ostracism, leaving the vacancy on the opposite side of the cluster.¹⁸ Once in any of these excited levels, the promoted atom and the vacancy can move about rather freely, including via exchanges of the promoted atom with others, giving rise to liquid-like behavior.

The 14-atom cluster does not exhibit sharply distinguished liquid-like and solid-like behavior. This is largely because the promotion energy to the first excited wells is significantly lower and the entropy is higher than for the 13-particle cluster. The reason is that an atom promoted from the icosahedral shell may be a nearest neighbor of the 14th atom, which resides on the surface of the icosahedron in the lowest energy geometry. Hence, low-energy excited wells are available to this cluster, and exchanges between surface and shell sites are much more facile than in the 13-particle cluster.

The question arises as to whether surface melting may occur in clusters. The answer is that it can in clusters large enough to support a solid core. The 13-atom cluster obviously cannot, but neither can the 33-atom cluster. The 55-atom cluster *does* exhibit surface melting, and it does this whether confined to a fixed volume as done by Nauchitel and Pertsin,²⁰ or put into a free volume; we have recently found the latter result. Furthermore, some of the other clusters slightly smaller and slightly larger than $N = 55$ also exhibit surface melting. However, one aspect of surface melting in clusters deserves special mention: it is not the outermost layer that melts, it is the *two* outermost layers.

This brings us to one other amusing aspect of the dynamics of clusters. One of the diagnostic tools most informative about whether a cluster is solid-like or liquid-like is a graph of the mean square displacement as a function of time. The slope of this curve is the diffusion coefficient. The simulated Ar₅₅ cluster shows its surface melting clearly by showing a definite positive slope in such a curve for the two outer layers, while the innermost layer shows a horizontal line with only a bit of noise. However, in a narrow band of energy just below the energy at which the surface melts, the outermost layer shows no mobility or diffusion, while the

next-outermost layer is almost as mobile as in the surface-melted state. These two situations are shown in Fig. 2; there, for illustrative purposes, the “normal” 55-atom cluster with a liquid surface and a solid core is illustrated by Cu₅₅, rather than Ar₅₅, to show the relative insensitivity of the surface melting result to potential. However, the “subsurface layer mobility” has only been found thus far in simulations of argon clusters.

The explanation of the “subsurface layer mobility” is this. The outermost layer of the 55-atom icosahedron is a set of 12 atoms atop the 12 of the inner shell of 12. The next-outermost layer consists of 30 atoms, one in the middle of each edge of the icosahedron. Hence, each of the outermost atoms rides in the center of a ring of five atoms in the next-outermost layer. Each atom in the next-outermost layer is in two such rings. Each ring can rotate relatively freely about the outermost atom it surrounds, leaving that atom almost unperturbed. But

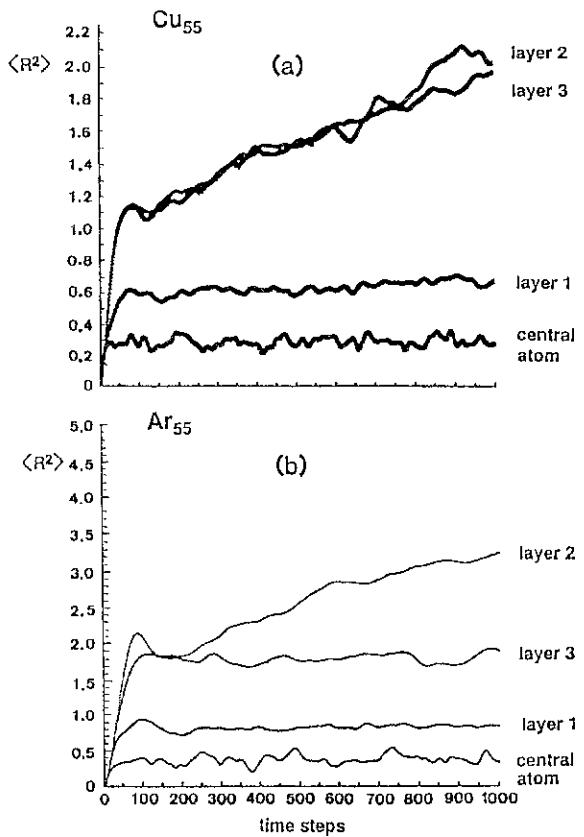


Fig. 2. Curves of mean square displacement of atoms in 55-particle clusters, with different shells plotted separately: the Cu₅₅ cluster, represented by an “embedded-atom” potential, under conditions where the surface is liquid and the core is solid (a); the Ar₅₅ cluster, represented by a Lennard-Jones potential, under conditions where the next-outermost (edge-centered) layer is mobile and all the others are near-rigid (b).

because any atom in the next-outermost layer can move with the rotation of either of two rings, it can find itself being permuted with other atoms of the same shell, eventually occupying all the sites of that shell with all the other members of the shell appearing in all possible positions as its neighbor. Hence the next-outermost layer can look liquid-like, yet leave all the rest of the 55-atom cluster solid-like!

SUMMARY AND OUTLOOK

The dynamics of particle motion in clusters exhibits a rich range of situations, including not only clear-cut solid- and liquid-like behavior and stiff solid-soft solid behavior. There are also cases of two-phase equilibrium within a cluster, e.g., solid interior and liquid surface, for a case in which the less dense liquid wets the more dense solid, and at least one example of a single mobile shell which is not the outermost.

Clusters are complex enough that it is undesirable to try to work with their potential surfaces in full detail. A direction that now looks attractive is a procedure in which the topology of the surface would be determined from a knowledge of the energies and geometries of the minima and of the saddles that link them. Here we give a little preview of work in progress. Using the kind of analysis presented for the topology of the six-particle cluster, one can find all the potential wells that can be reached at any fixed energy. Then, by making a harmonic approximation for the normal modes in each well, one can estimate the hyperarea available in each well, count the number and kind of reachable wells, and thereby estimate the total available area, a number whose logarithm is proportional to the entropy of the isoergic system at that energy. This has been the information most difficult to obtain in attempts to derive thermodynamic properties from knowledge of potential surfaces. With this information, it will be possible to make at least approximate estimates of the free energies of clusters of any energy, whether they explore all or only part of their formally available configuration space. From this information, we shall be able to determine the kinds of "phase changes" that clusters can undergo, of the conditions of energy or temperature at which those transformations will occur, and of the energies and entropies associated with those transformations. From there, we can perhaps begin to connect our inklings of understanding of phase changes with the chemical changes that reacting clusters undergo.

Acknowledgment. This research was supported by a Grant from the National Science Foundation.

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