

The dynamics of matrix trapping. II. Simple spherical species trapped in an argon matrix

Astrid J. Cruz and Gustavo E. López

Citation: *The Journal of Chemical Physics* **104**, 4301 (1996); doi: 10.1063/1.471159

View online: <http://dx.doi.org/10.1063/1.471159>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/jcp/104/11?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Ultrafast quantum dynamics and resonance Raman spectroscopy of photoexcited I₂\(B\) in large argon and xenon clusters](#)

J. Chem. Phys. **104**, 9332 (1996); 10.1063/1.471678

[Interrogation and control of condensed phase chemical dynamics with linearly chirped pulses: I₂ in solid Kr](#)

J. Chem. Phys. **104**, 6497 (1996); 10.1063/1.471370

[Poling dynamics and investigation into the behavior of trapped charge in poled polymer films for nonlinear optical applications](#)

J. Chem. Phys. **104**, 6834 (1996); 10.1063/1.471350

[Manybody potentials and dynamics based on diatomic molecules: Vibrational frequency shifts in Ar_n HF \(n=1–12,62\) clusters](#)

J. Chem. Phys. **104**, 5510 (1996); 10.1063/1.471790

[The dynamics of matrix trapping. I. Deposition of a pure argon matrix](#)

J. Chem. Phys. **104**, 4294 (1996); 10.1063/1.471239



The dynamics of matrix trapping. II. Simple spherical species trapped in an argon matrix

Astrid J. Cruz and Gustavo E. López

Department of Chemistry, University of Puerto Rico at Mayagüez, Mayagüez, Puerto Rico 00680

(Received 28 March 1995; accepted 15 December 1995)

The technique of matrix isolation has extensively been used to study reactive species which are isolated in an inert host matrix, preventing in this way diffusion or reaction of the trapped species. The present study is concerned with the theoretical description of matrix trapping dynamics. The most common experimental technique of matrix deposition, slow spray on, is simulated using classical molecular dynamics. The systems considered are simple spherical species trapped in argon. The effect of the size of the trapped species on the structure of the matrix and the nature of the trapping site is investigated. © 1996 American Institute of Physics. [S0021-9606(96)50911-0]

I. INTRODUCTION

Matrix-isolation (MI) spectroscopy has been extensively developed as a mean of studying reactive molecular species such as free-radicals, which in the gas phase have extremely short lifetimes. In its most commonly used form, MI entails the preparation of a rigid sample in which the molecular species of interest is isolated and effectively immobilized in the inert host lattice or matrix. The inertness of the matrix will prevent loss of the trapped species by reacting with the host material. As stated in our previous paper,¹ the structural nature of the matrix/trapped-species system is seldom known. The primary thrust of the present investigation is to contribute to the fundamental understanding of the structure of the trapping site of simple spherical species trapped in a rare gas matrix. The dominant conventional view, well illustrated in the book of Beat Mayer,² is that the matrix trapping sites correspond to rare gas solid substitutional vacancies. For an impurity molecule of size comparable to the rare gas atom, a single substitutional vacancy provides the site for the trapped molecule. On the other hand, for a nonspherical impurity, the removal of a cluster of adjacent rare gas atoms provides the trapping site. In the latter case, the removed rare gas atoms might mimic the shape requirements of the trapped molecules. Thus a spatially compatible contiguous cluster of the rare gas atoms plays a major role in speculating about trapping sites for impurity molecules. In this picture the matrix relaxation about the trapping entity is a secondary feature. The modification of the matrix material is limited to a short-range local environment about the impurity. However, this picture does not consider any major long-range reorganization of the host rare gas material.

There is little experimental information to elucidate the effects of the impurity molecule on the structure of the host matrix material. A major obstacle to an experimental contribution is the low concentration of the trapped molecule which is present in such dilute amounts that the usual considerations of alloys substances is of questionable value. We can, however, look to parallels for guidance, but with caution. There are EXAFS studies³ of rare gas alloys which indicate a random substitutional structure with relatively small distortions, but these alloys have compositions well out

of the range of matrix trapping. There are well-known experimental crystal structure studies by Knozinger *et al.*⁴ on rare gas matrices doped with a varying percentage of simple molecular impurities. In these studies the crystal structure of argon undergoes a distortion. More extensively, but perhaps only generally useful, are many studies of metal alloys or solid solutions. This work is well summarized by Barrett and Massalski.⁵ A number of empirical models have been developed—most notable the Hume–Rothery rules and many others—which attempt to correlate the alloys structure with the relative atomic sizes (and other properties) of the alloy components. These Hume–Rothery rules, however, are applied to metal alloys in which the amount of the lesser component is present in greater concentration than is common to the matrix trapping case. The general lore of crystallization also takes note of pronounced changes in crystallization when impurities are incorporated, but detailed specifics are often unknown. Thus there is very little specific guidance in consideration of the structure of matrix isolation materials involving complex impurities.

From a theoretical point of view, very little specific information is available with regard to the structural problem in MI. Some earlier studies include classical lattice dynamics calculations⁶ where the system is treated as a crystal lattice with the impurity in a substitutional site. However, this method presents the limitation that it can only be applied to well-defined configurations of the crystal, i.e., glassy material or even considerable disordered matrix-isolated cases require special treatment.

Recently, Monte Carlo (MC) and molecular dynamics (MD) simulations⁷ have been used to study several matrix-isolated systems, i.e., rare gas atoms and small diatomic molecules trapped in rare gas solids. These simulations focused primarily on the structural and dynamical behavior of these systems. However, the question of the specific nature of the MI system was not addressed in these studies. Basically, the simulation started from a preformed system in which the matrix was assumed to be in a fcc lattice with the impurity trapped in a substitutional site.

Recently, Fraenkel and Haas⁸ presented a study of trapping of a guest molecule in a rare gas matrix using MD

techniques. Their results suggest a strong dependence on the preparation of the system and on the size of the trapped species. The main difference between their and our study is that not only we considered various small impurities trapped in argon but also our method to dissipate the excess energy of the incoming atoms in the deposition process is more efficient. In their work the energy dissipation is achieved by periodically rescaling the velocities of all the atoms in the system. This procedure can in principle disrupt the dynamical evolution of the system. As discussed in Paper I, our dissipation process is based on rescaling only a subset of atoms in the system in the same fashion that other studies,⁹ which has considered the dissipation of energy in solids, have done it. As discussed in Sec. IV our results are in qualitative agreement with the results obtained by Fraenkel and Hass.

It is the primary purpose of this theoretical work to probe the effects of the impurity size in the matrix trapping process—the additional effects of molecular shape and chemical specificity are undoubtedly important but their investigation must follow the present, more basic, study. Specifically, the following points are to be addressed; where is the trapped molecule in the matrix, and how should the entire material be characterized, i.e., crystalline, amorphous, or polycrystalline? Also of interest is the nature and degree of the matrix distortion/response near the trapped molecule. The matrix/impurity systems considered are Ne/Ar_N, Xe/Ar_N and SF₆/Ar_N. The simulational techniques described in Paper I for the vapor deposition of the pure matrix are applied to the above systems. The influence of the trapped species on the matrix will be characterized by comparison with the results presented in Paper I for the pure matrix.

In Sec. II we present the details of the theoretical method. In Sec. III results and discussion are presented. Finally, in Sec. IV we summarize our results.

II. THEORETICAL MODELS

The simulational techniques used are those studied and discussed in Paper I. We will present only the pertinent details for the systems considered here. The interparticle interaction between Ar–Ar and Ar–X (where X=trapped species) has been represented by 12-6 Lennard-Jones (L-J) potentials. In the present work, we have used these simple potentials rather than more “realistic” potentials¹⁰ for several reasons. First, it is well established that satisfactory results for liquid and solid rare gases can be obtained with this level of approximation. Moreover, the simplicity of the potential makes the calculation of forces, which is the bulk of the computational labor in MD simulations, simple and relatively fast. In addition, at this level of investigation, we are less interested in detailed specific systems, but rather in probing the general aspects of the matrix/trapped specie system, i.e., how the general shape and size of the isolated species affects the structure of the matrix. In the last place, uniformly high quality potentials for a pure substance and especially for different unlike pairs are not easily available, i.e., it is not sensible to use a superb potential for Ar–Ar interaction and rather

TABLE I. Parameters for the 12-6 L-J potential between like species and for the 9-3 L-J potential between rare-gases and a rigid graphite surface.

Atom	σ (Å)	ϵ (K)	s_0 (Å)	ϵ_s (K)
Ne	2.78	36.46	1.59	439.69
Ar	3.405	119.8	1.91	1155.0
Xe	3.75	230.96	2.25	1977.5
SF ₆	4.52	468.1	2.34	4205.3

poorer ones otherwise. Therefore, at the present level we use a generic L-J potentials which takes into account the general features of simple atoms and molecules. Strictly speaking we consider the matrix isolation of L-J systems. When the potential parameters were not available for the binary systems, e.g., Ar–Ne, Ar–Kr, and Ar–SF₆, the simple combining rules of Berthelot–Lorentz were used.¹⁰ The interaction potential between Ar or X and the rigid substrate is described by a 9-3 L-J potential.¹¹ Table I contains a summary of the atom–atom and surface-adsorbed particles parameters used in this work.

To simulate the slow spray on deposition and the pulsed deposition techniques, the MD procedure described in Paper I was used. Namely, the dynamics of the system starts with one or two seeded layers and the addition of one atom (or cluster of atoms) and continues until all the N matrix atoms and n impurity atoms are introduced. After the introduction of all atoms, the simulation is continued until equilibrium is attained.

III. RESULTS AND DISCUSSION

A. Deposition of matrix/trapped-specie system

The first system considered in this study was composed of one neon atom trapped in a 499-atoms argon matrix, which corresponds to a ratio commonly used in MI experiments. Neon is a spherical atom which is approximately 0.8 times smaller than argon. Therefore, in this particular case, we are interested in the effect that trapping a relatively small spherical species (relative to argon) will have on the host material.

The initial conditions for this simulation are the following: An initial random layer of argon atoms is placed on top of the graphite surface at a temperature of 10 K. As in Paper I, the velocities of the atoms on the seeded layers are rescaled every 10 Δt to maintain the desired temperature. In this particular simulation, we have used the slow spray-on technique with a rate of deposition of 1 atom every 5 Δt . Atoms are introduced one at a time as in the pure argon study except that a neon atom is introduced as the 200th atom.

The results for this system are shown in Figs. 1 through 3. First, it is observed from Fig. 1 that the y – z projection of the atomic positions shows a layer-by-layer formation similar to the one observed for pure argon. The same type of structure is apparent from Fig. 2 which shows the x – y projection for the first five layers and the corresponding two-dimensional pair distribution function (2D PDF). The neon atom is located in the fourth layer and specifically in an argon lattice substitutional site. All layers, including the

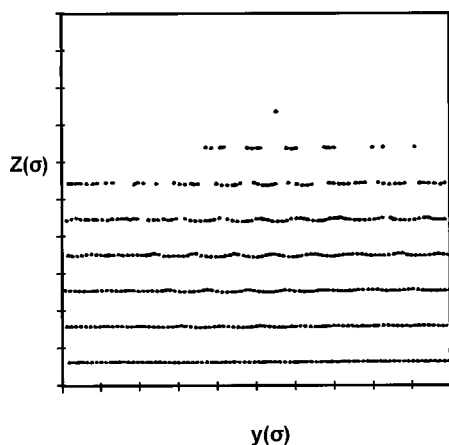


FIG. 1. The y - z projection of the atomic positions for the $\text{Ar}_{499}/\text{Ne}$ system deposited onto a flat graphite surface.

fourth one, show the usual close-packing structure obtained for pure argon. Moreover, in all cases the 2D PDF shows little distortion with respect to perfect close-packing structure previously obtained for the pure matrix.

To characterize the distortion around the neon atoms in a more quantitative way, the distribution function of distances, $P(r)$, is calculated for the neon atom and a nearby argon atom. Figure 3(a) shows $P(r)$ for neon. The typical shape of a close-packing structure is obtained, but with all peaks slightly shifted toward smaller r values, relative to the perfect close-packing structure. This is due to contraction of the nearest neighbors of neon. Calculation of $P(r)$ for one of neon nearest neighbors [Fig. 3(b)] shows the same small displacement of the peaks. $P(r)$ was also calculated for second and third nearest neighbors of neon and a nearly undisturbed perfect close-packing was observed.

Next, we considered one xenon atom trapped in an argon matrix composed of 599 atoms. Xenon is approximately 1.1 times larger than the argon atom. The simulational conditions used are the same as for neon.

The results for the Xe/Ar_N system are shown in Figs. 4 through 6. Figure 4 shows a layer-by-layer formation similar to all previous cases. Figure 5 shows the atomic positions in the x - y plane for the first five layers, and the corresponding 2D PDF. The xenon atom is located in a substitutional site in the third layer. All layers show a closed-packed structure with a three dimensional hcp arrangement and even in the third layer where the impurity was trapped, the 2D PDF shows little distortion with respect to the perfect close-packing.

Figure 6 shows $P(r)$ for the Xe atom and for a nearby argon atom. A more pronounced distortion is now observed for the nearest-neighbor atoms of xenon—all peaks have been shifted and distorted toward larger r values. Hence, the matrix has been expanded around the impurity, which is larger in size than the matrix atoms, in order to accommodate it. On the other hand, if $P(r)$ is computed for a second nearest-neighbor argon atom, the usual nearly undisturbed close-packing arrangement is observed. Hence, it can be con-

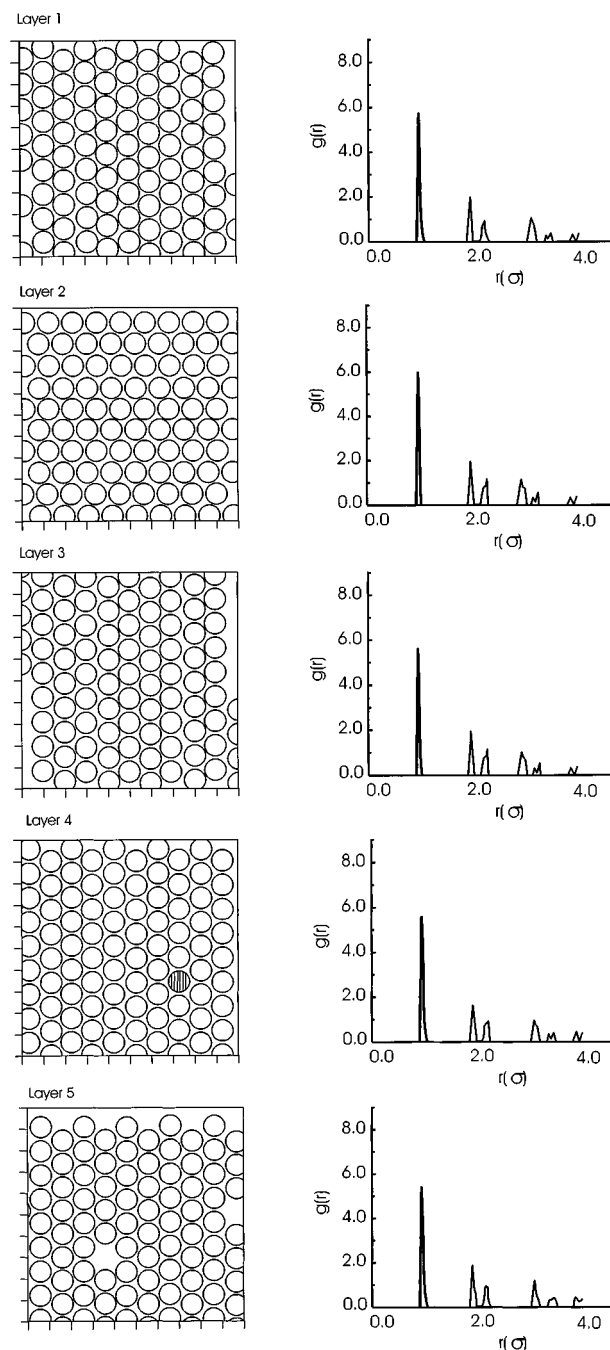


FIG. 2. The x - y projection of the atomic positions and the two-dimensional pair distribution function for the first five layers of the $\text{Ar}_{499}/\text{Ne}$ system.

cluded that the xenon impurity causes a localized distortion of the argon matrix, namely, only the first nearest neighbors are being significantly affected.

The third case we have considered in this study is the simulation of slow spray-on deposition of one SF_6 molecule and 500 argon atoms. The SF_6 is an octahedral molecule which can be viewed as a large spherical species—approximately 1.3 times larger than argon. In this particular case we are interested in the effects that a very large species

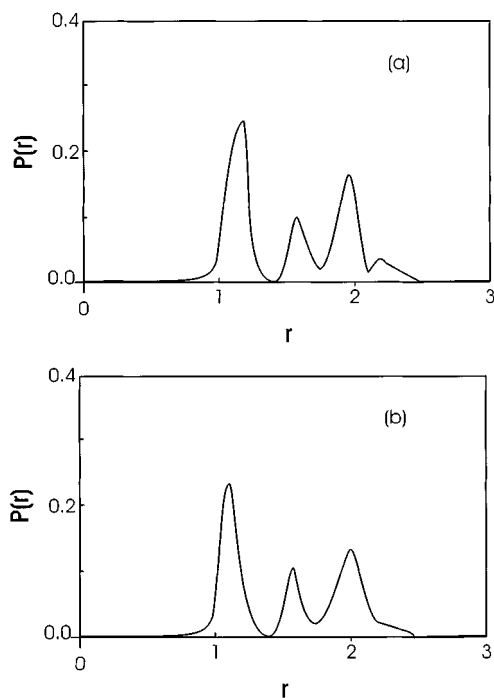


FIG. 3. (a) Distribution function of distance, $P(r)$, for Ne in the $\text{Ar}_{499}/\text{Ne}$ system. (b) Same as (a) but for a second nearest neighbor of neon.

would have on the host material. The initial conditions for this simulation are exactly the same as the previously described for the spray-on simulations of Ne/Ar_N and Xe/Ar_N systems.

The results for the SF_6/Ar_N system are presented in Figs. 7 through 9. At first glance, the y - z perspective of the atomic positions shows the expected layer-by-layer type of formation obtained in the previous cases (Fig. 7). However, careful examination of Fig. 7 reveals that several atoms are accommodated between layers, a phenomena that was not previously observed. In particular, it can be seen that the SF_6 is located between the second and third layer. Examination of the PDF shows a reduction of long-range ordering.

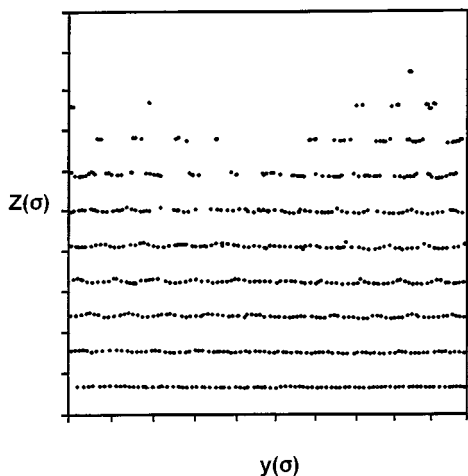


FIG. 4. Same as Fig. 1 but for the $\text{Ar}_{599}/\text{Xe}$ system.

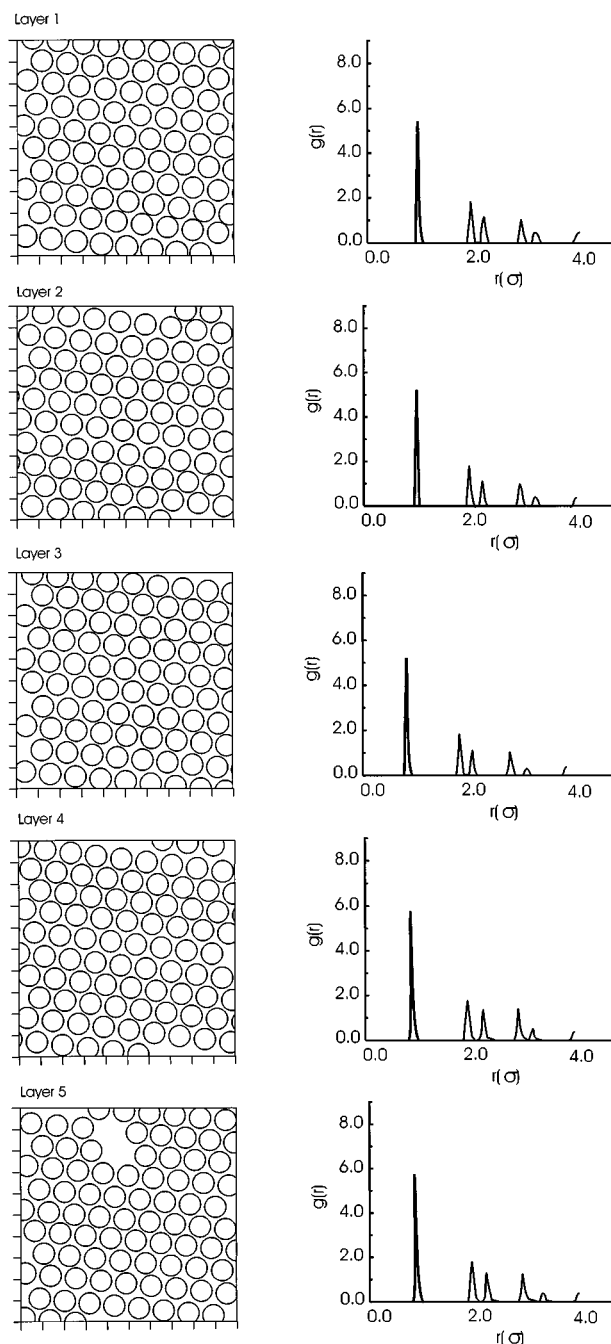


FIG. 5. Same as Fig. 2 but for the $\text{Ar}_{599}/\text{Xe}$ system.

To have a clearer idea of the effects caused by the trapped SF_6 molecule, the atomic position of the first eight layer and the corresponding 2D PDF are presented in Fig. 8. The matrix is considerably distorted in the presence of the SF_6 impurity. There is a three atom wide section in the z direction of the matrix that has a different packing arrangement than the rest of the matrix, i.e., a slightly tilted section in the z direction. Moreover, the SF_6 molecule is trapped between the second and third layer of this vertical slab. Simultaneously, a number of voids can be observed in the first two layers of this section. The fourth and fifth layer do not show voids, but the different packing of atoms in the tilted

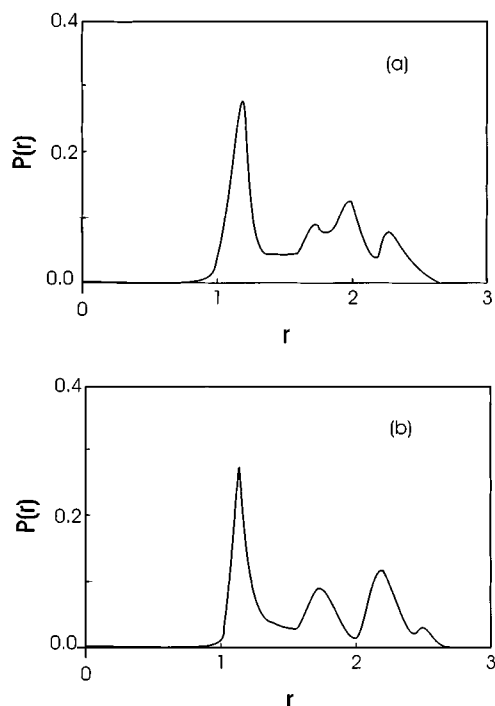


FIG. 6. (a) Distribution function of distance, $P(r)$, for Xe in the $\text{Ar}_{599}/\text{Xe}$ system. (b) Same as (a) but for a first nearest neighbor of xenon.

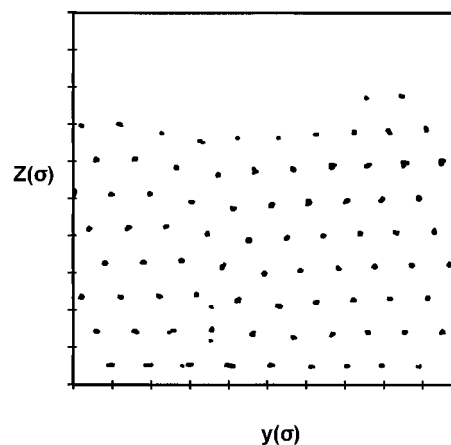


FIG. 7. Same as Fig. 1 but for the $\text{Ar}_{599}/\text{SF}_6$ system.

portion of the cell is clearly visible. The 2D PDF of each layer shows major deviations from the one corresponding to the usual close-packing structure. In particular, several new peaks are present, which probably correspond to the atoms in or around the vertical tilted slab.

Figure 9 shows the distribution function of distances for several particular atoms in this system. Specifically, Fig. 9(a) shows $P(r)$ for a nearest neighbor of SF_6 and its form does not correspond to any type of simple crystalline structure. All peaks have coalesced, which means that there are atoms at practically all possible values of r , hence overall periodicity

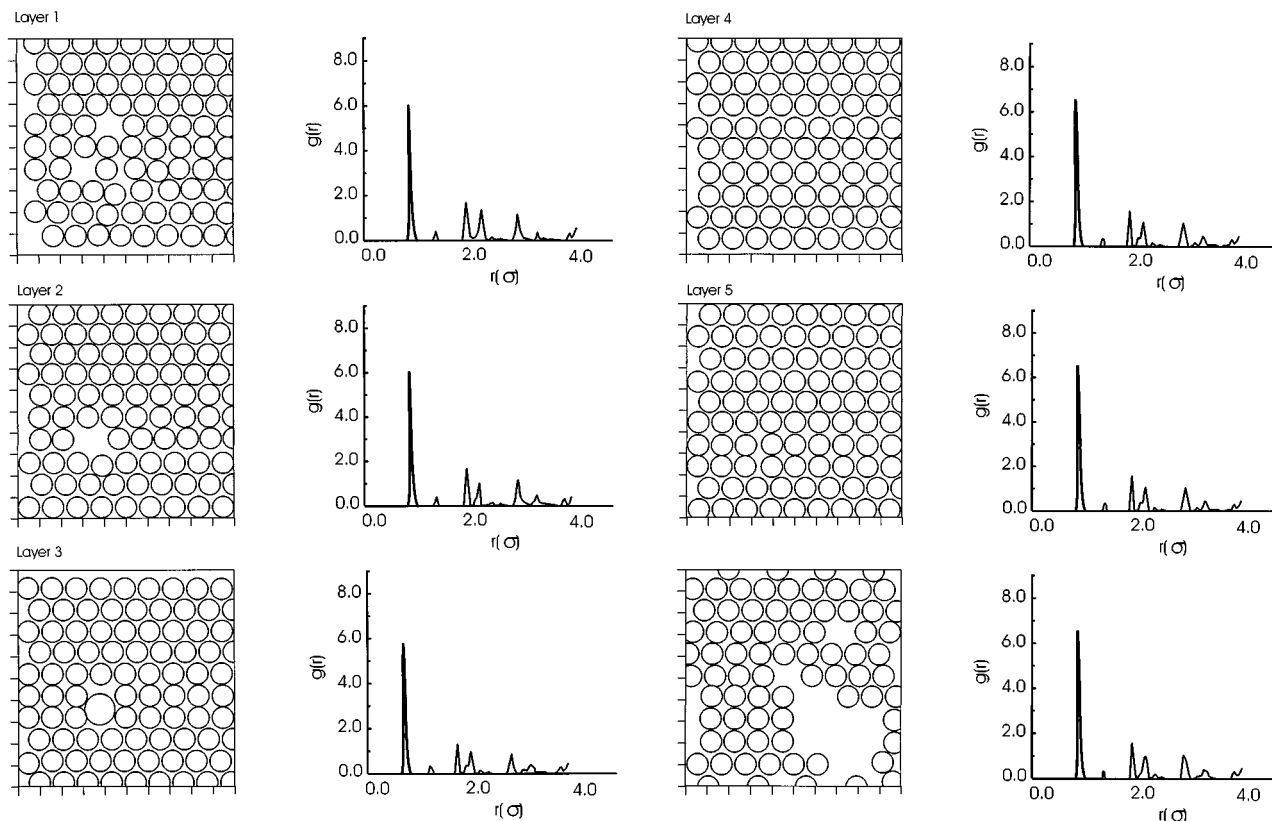


FIG. 8. Same as Fig. 2 but for the first six layers of the $\text{Ar}_{599}/\text{SF}_6$ system.

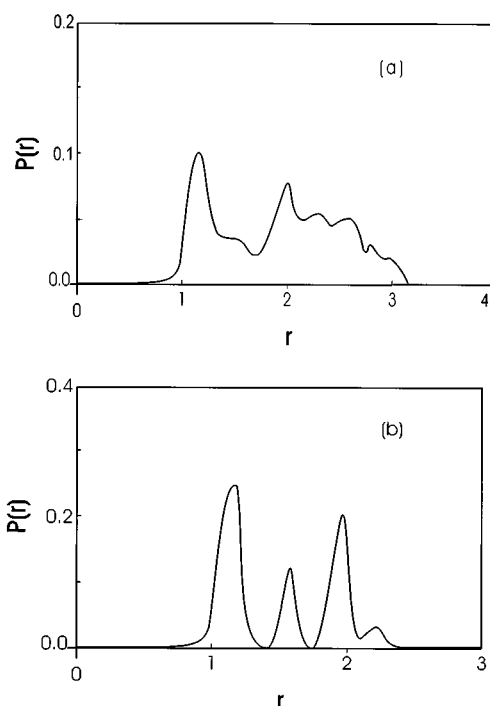


FIG. 9. (a) Distribution function of distance, $P(r)$, for SF_6 in the $\text{Ar}_{599}/\text{SF}_6$ system. (b) Same as (a) but for a first nearest neighbor of xenon.

(long range in the cell) has been lost. However, $P(r)$ for an argon atom far from the SF_6 molecule (outside of the tilted region in the simulational box) exhibits the usual close-packing shape [Fig. 9(b)].

In order to ensure that the results obtained are not artifacts caused by the small size of the cell, a simulation was made using the same conditions as in the previous one, but in a $16\sigma \times 16\sigma \times 16\sigma$ cell. Qualitative agreement was obtained with respect to previous smaller cell, namely, the SF_6 is trapped between two layers and long-range distortion is observed. A large rectangular simulational box was also used, and the results obtained were unchanged.

For this particular system, the idea of describing the nature of the trapping site is vague. Namely, the SF_6 is not simply accommodated in a normal substitutional site, nor in an interstitial site because interstitial sites can not be easily defined in this highly distorted system. Perhaps the best way of describing the trapping site is as a "cage," i.e., the matrix atoms form a cage around the trapped species and reveal no particular crystalline arrangement. More interesting is the fact that the simple idea of removing matrix atoms from the crystal in order to accommodate the trapped species does not seem valid for this particular system. Contrary to the previous cases where only local distortion was observed, SF_6 caused a long-range effect in the crystal, i.e., the entire material (500 atoms) is affected by the presence of the impurity. However, a certain degree of order is preserved within the material—it could not be described as a glass, for example.

Several simulations were performed in order to elucidate if the observed long-ranged distortions in the previous results are a consequence of the strong interaction potential between

TABLE II. Parameters for the MD simulation of impurity of variable size trapped in argon.

Run	σ (Å)	ϵ (K)	s_0 (Å)	ϵ_s (K)
1	3.625	189.5	1.996	1765.1
2	3.845	259.3	2.082	2375.1
3	4.065	329.0	2.168	2985.2
4	4.285	398.8	2.254	3595.2
5	4.520	468.5	2.34	4205.3

the SF_6 molecule and the graphite surface. For this purpose the $\text{SF}_6/\text{graphite}$ potential was decreased to approximately the value of the $\text{Ar}/\text{graphite}$ potential. No appreciable changes are observed by this scaling of the surface potential—the same long-ranged distortion of the matrix was obtained which suggest that it is not caused by the interaction between the graphite and the trapped species, but rather by the size effect that the impurity has on the matrix.

B. MD simulation of impurity of variable size trapped in argon

The explicit effects of increasing the ratio of $\sigma(X)/\sigma(\text{Ar})$, where $\sigma(X)$ is the radius of the impurity X, on the structure of the matrix is approached in another way in this section. From all previous results it is apparent that the distortion of the matrix depends to a great extent on the size of the impurity. For an impurity molecule which is approximately the same size as the matrix atoms [$\sigma(X)/\sigma(\text{Ar}) \approx 1$] the distortion is very localized, whereas for larger impurities the distortion of the matrix can be significant. Therefore, it would be useful to perform a sequence of MD simulations where the distortion of the deposited matrix is monitored as a function of the size of the impurity. This kind of simulation does not parallel any real experimental technique used in matrix trapping. However, it provides useful information related to the correlation between the impurity size and the extent and nature of the matrix deformation.

These MD simulations begin with a pure solid argon matrix, formed by a slow spray-on deposition simulation, composed of 600 atoms in contact with the 9-3 L-J surface. Then the size of the 200th argon atom, which is approximately localized in the center of the matrix, is increased by a factor of 0.22. Correspondingly, all the other potential parameters are linearly scaled. The parameter $\epsilon_{\text{Ar-Ar}}$, $\epsilon_{\text{Ar-X}}$, and all the parameters representing the interaction between the graphite and the matrix/impurity are linearly increased as to obtain the SF_6 parameters after five simulations. The parameters generated by this method are summarized in Table II. The remaining initial conditions are similar to the previous slow spray-on simulations. In all cases 2000 Δt between increments of the potential parameters were enough to allow the new system to reach equilibrium. The initial configuration on each run comes from the previous equilibrated simulation.

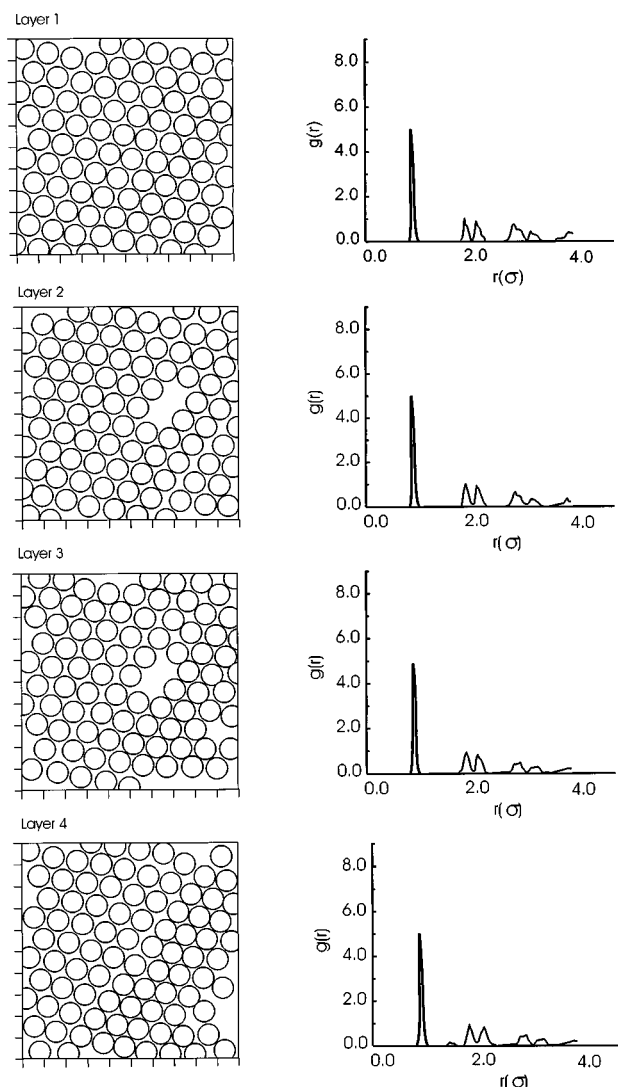


FIG. 10. The x - y projection of the atomic positions and the two-dimensional pair distribution function for the first four layers of the fourth run in the MD simulation of impurity of variable size trapped in argon.

The first three simulations show results in agreement with our previous results for xenon in argon. Namely, the matrix is slightly distorted due to the spherical impurity, which is trapped in a substitutional site of the argon matrix. In all three cases the distortion is local and becomes more pronounced as the size of the impurity is increased. The argon matrix conserves the layer-by-layer type of formation with a nearly perfect hcp structure, similar to the one obtained for pure argon.

In the fourth run the matrix presents long range deformation as shown in Fig. 10 by the x - y projection of the first four layers and the corresponding 2D PDF. The impurity is trapped between two layers and considerable distortion is observed in the region where the impurity is trapped. Contrary to the first three simulations, the structure of the entire material is affected by the trapped species. It is important to notice that the fourth run corresponds to a species which is 0.22 smaller than SF_6 . The fifth run, which corresponds to SF_6 , gives results which are in qualitative agreement with

the results obtained using the slow-spray on technique.

IV. CONCLUSIONS

In the MI technique it is of utmost importance to understand the nature of the trapping site as well as the structure of the host material. In the present studies we have performed simulations of the two most popular matrix deposition techniques: slow spray-on and pulsed deposition. We have focused on the effect that the size of the trapped specie has on determining the location of the trapping site and the degree of the matrix distortion. The results demonstrate that the presence of a spherical trapped species of comparable size with the matrix atoms causes no significant change on the structure of the host matrix, i.e., the argon/X, where $X = \text{Xe}$ or Ne forms a single crystal with hcp structure under the simulation conditions used in this work. The impurity is trapped in a substitutional site, which is consistent with simple physical size considerations. Obviously, in the case of an argon matrix, an interstitial site is too small to accommodate neon without causing major distortion, hence being energetically unfavorable.

A spherical impurity as SF_6 , which is considerably larger than the matrix atoms, causes remarkably different results. A major structural reorganization of the matrix is observed. Instead of accommodating the impurity in several adjacent argon sites, the matrix will rearrange its structure in order to minimize the configurational energy of the system. The nature of the trapping site is unclear owing to the fact that the material cannot be described as a single crystal although several regions with crystalline character are observed. Our results are in agreement with the work by Fraenkel and Haas⁸ where a dislocated matrix is obtained when the guest molecule is SF_6 . Also, as in our work, Fraenkel and Haas demonstrated that if the system is properly equilibrated, a nearly defect-free matrix is obtained.

ACKNOWLEDGMENTS

A.J.C. acknowledges support from SEED MONEY Program. G.E.L. acknowledges financial support from the Research Corporation.

¹ A. J. Cruz and G. E. Lopez, J. Chem. Phys. **104**, 4294 (1996).

² B. Meyer, *Low Temperature Spectroscopy; Optical Properties of Molecules in Matrices: Mixed Crystals and Frozen Solutions* (American Elsevier, New York, 1971).

³ W. Malzfeldt, W. Niemann, P. Rabe, and R. Haensel, in *EXAFS and Near Edge Structure III*, edited by K. O. Hodgson, B. Hedman, and J. E. Penner-Hahn (Springer, Berlin, 1984); W. Malzfeldt, W. Niemann, P. Rabe, and R. N. Schwentner, in *EXAFS and Near Edge Structure*, edited by A. Bianconi, I. Incoccia and S. Stipcich (Springer, Berlin, 1983).

⁴ E. Knozinger, W. Schuller, and W. Langel, Faraday Discuss. Chem. Soc. **86**, 285 (1988); E. Knozinger and R. Wittenberg, J. Am. Chem. Soc. **105**, 2154 (1983).

⁵ C. S. Barrett and T. B. Massalski, *Structure of Metals; Crystallographic Methods, Principles and Data*, (Pergamon, New York, 1980).

⁶ See for example, H. J. Jodl, in *Chemistry and Physics of Matrix-Isolated Species*, edited by I. Andrew and M. Moskovits (Elsevier Science, New York, 1989).

- ⁷S. Nosé and M. L. Klein, *Can. J. Phys.* **60**, 1365 (1982); M. E. Fajardo, *J. Chem. Phys.* **98**, 119 (1993); J. A. Boatz and M. E. Fajardo, *ibid.* **101**, 3472 (1994).
- ⁸R. Fraenkel and Y. Hass, *J. Chem. Phys.* **100**, 4324 (1994).
- ⁹H. C. Andersen, *J. Chem. Phys.* **72**, 2384 (1980); M. E. Riley, M. E. Coltrin, and D. J. Diestler, *ibid.* **88**, 5934 (1988); A. E. DePristo and H. Metiu, *ibid.* **90**, 1229 (1989).
- ¹⁰R. A. Aziz, in *Inert Gases*, edited by M. L. Klein (Springer, Berlin, 1984).
- ¹¹W. A. Steele, *Interaction of Gases with Solid Surfaces* (Pergamon, Oxford, 1974).