

## Computer simulation of site formation for Na atoms trapped in Ar and Xe solids

L. C. Balling and J. J. Wright

Citation: *The Journal of Chemical Physics* **81**, 675 (1984); doi: 10.1063/1.447702

View online: <http://dx.doi.org/10.1063/1.447702>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/jcp/81/2?ver=pdfcov>

Published by the [AIP Publishing](#)

---

### Articles you may be interested in

[Electron traps in solid Xe](#)

*Low Temp. Phys.* **35**, 335 (2009); 10.1063/1.3117964

[Monte Carlo simulations of the structures and optical absorption spectra of Na atoms in Ar clusters, surfaces, and solids](#)

*J. Chem. Phys.* **101**, 3472 (1994); 10.1063/1.467532

[Classical Monte Carlo simulations of relaxed trapping site structures in Li atom doped solid Ne](#)

*J. Chem. Phys.* **98**, 119 (1993); 10.1063/1.464660

[Multiple trapping sites and symmetry splitting in cryogenic matrices: Infrared spectroscopy of HCN in Ar, Kr, and Xe](#)

*J. Chem. Phys.* **82**, 1255 (1985); 10.1063/1.448446

[Total Cross Sections for Formation of Ions from CsBr by Collision with Ar, Xe, and NaBr\(Ar\)](#)

*J. Chem. Phys.* **57**, 4742 (1972); 10.1063/1.1678145

---



# Computer simulation of site formation for Na atoms trapped in Ar and Xe solids<sup>a)</sup>

L. C. Balling and J. J. Wright

*Department of Physics, University of New Hampshire, Durham, New Hampshire 03824*

(Received 10 February 1984; accepted 10 April 1984)

Existing data on Na-rare gas and rare gas-rare gas dimer potentials have been used in a computer program to simulate the formation of trapping sites for Na atoms isolated in Ar and Xe solids. The dimer potentials have also been employed to calculate the matrix perturbed energy-level structure of the Na atoms to obtain theoretical wavelengths for absorption and emission bands corresponding to the sites formed in the simulations. The results support the view that the stable blue-shifted absorption band of Na in Ar and Xe is due to a substitutional site and that the thermally unstable unshifted band results from amorphous sites evolving from two-atom vacancies. In all cases, the simulated sites exhibited a local symmetry sufficient to produce a degeneracy in the perturbed Na 3*p* state levels.

## I. INTRODUCTION

Experimental studies of the optical spectra of matrix-isolated alkali atoms<sup>1-4</sup> and of group II atoms<sup>5</sup> clearly indicate that these atoms can be trapped in a number of different types of sites of varying thermal stability, and the configuration of these sites is still an open question. Previous theoretical calculations<sup>6-8</sup> of the absorption- and emission-band wavelengths of matrix-isolated alkali atoms have been based on simple trapping-site models, in which the alkali atom is located in a one-, two-, three-, or four-atom vacancy in an essentially regular rare gas crystal.

Given the weakness of the Van der Waals binding in rare gas solids, it is reasonable to question whether or not an alkali atom can be trapped in a substitutional site, as the equilibrium separation between a ground-state alkali atom and a rare gas atom is, in general, substantially larger than the internuclear distance in a rare gas crystal. Furthermore, if the alkali is trapped in sites of larger volume, one must also question the validity of simple undistorted multiatom vacancies as site models.

One advantage of considering matrix-isolated alkali atoms is that the ground-state and excited-state interactions of alkali atoms with rare gas atoms have been extensively studied, both theoretically and experimentally, and alkali-rare gas dimer potentials are available in the literature. In a previous paper<sup>8</sup> we showed how existing experimental and theoretical alkali-rare gas dimer potentials can be used to calculate the perturbed energy-level structure of alkali atoms in model trapping sites, and we obtained theoretical absorption- and emission-band wavelengths for these sites.

It is the purpose of this paper to show how these dimer potentials, in conjunction with dimer potentials for the rare gas-rare gas interaction, can be used to simulate the growth of trapping sites for alkali atoms in rare gas solids and to simulate the motional relaxation of the atoms in the site in response to optical excitation of the alkali atom. We report here computer simulations of trapping-site formation for Na atoms in Ar and Xe matrices, simulations of motional relaxation in each site, and calculations of the corresponding ab-

sorption and emission wavelengths associated with the Na 3*s*-3*p* transition.

In Sec. II we describe our method for simulating matrix growth and motional relaxation. In Sec. III we present our results for Na in Ar and Xe and compare them with experiment and with other theoretical analyses.

## II. CALCULATIONAL METHOD

For both the simulation of matrix growth and the calculation of spectral wavelengths we require expressions for the potential energy of the Na atom interacting with an arbitrary number of rare gas atoms. As was shown in Ref. 8, one can express this potential in terms of Na-rare gas dimer potentials available in the literature. The calculation is an exercise in first-order degenerate perturbation theory. For the Na-Ar and Na-Xe dimer potentials we used existing experimental and theoretical potentials reported in Refs. 9-12.

### A. Matrix deposition

For ground-state Na atoms, the situation is particularly simple, because the perturbation theory result shows that the  $X^2\Sigma$  Na-rare gas potentials can be taken to be pairwise additive,<sup>8,13</sup> as is the case for the rare gas-rare gas interaction. This means that the total potential energy for a system of a Na atom and an assembly of *N* rare gas atoms can be written as

$$V(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_n, \mathbf{R}) = \sum_{i < j}^N V_{ij}(\mathbf{r}_i, \mathbf{r}_j) + \sum_i^N U_i(\mathbf{r}_i, \mathbf{R}), \quad (1)$$

where  $U_i$  is the  $X^2\Sigma$  potential energy of a Na atom at  $\mathbf{R}$  interacting with a rare gas atom at  $\mathbf{r}_i$  and  $V_{ij}$  is the interaction potential between two rare gas atoms at positions  $\mathbf{r}_i$  and  $\mathbf{r}_j$ . For  $V_{ij}$  we used the empirically determined rare gas interaction potentials reported in Refs. 14 and 15 for Ar and Xe, respectively.

The computer simulation of trapping-site formation relied on calculations of  $V$  as a function of the alkali and rare gas atom positions. The computer program allowed rare gas atoms and a Na atom to impinge at random upon a fcc rare gas crystal substrate, as shown schematically in Fig. 1. The rare gas atoms and the alkali atom then relaxed to equilibri-

<sup>a)</sup> This work was supported by NSF.

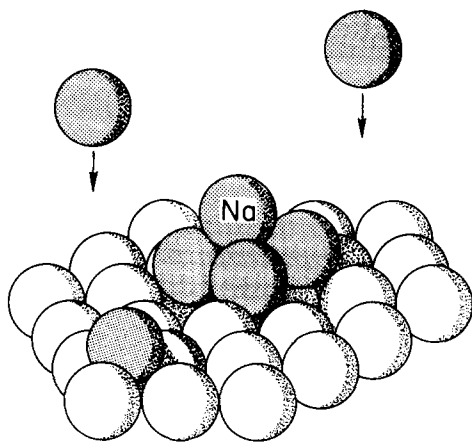


FIG. 1. A schematic representation of the computer-simulated deposition of a Na atom and rare gas atoms on a fixed substrate of 27 rare gas atoms arranged to form the (111) plane of an fcc crystal. The Na atom and all rare gas atoms, with the exception of the original 27, are allowed to move to new equilibrium positions as each new rare gas atom or group of atoms comes in.

um positions by following the gradient in the potential which each atom saw until the system's total potential energy  $V$  was minimized. The effect of thermal motion of the atoms was simulated by a simple device in the program which carried atoms over small potential bumps, thus avoiding pseudominima.

The crystal substrate consisted of 27 rare gas atoms which remained fixed in positions corresponding to the (111) plane of an fcc lattice. This arrangement of the substrate atoms, which also corresponds to the basal plane of an hcp structure, was chosen because of the tendency<sup>16,17</sup> of inert-gas crystallites to grow with this plane parallel to the supporting substrate.

The simulation program brought in rare gas atoms individually and in groups. Each incoming atom approached the matrix until the atom's contribution to the system's potential energy went positive and equalled the thermal energy of a 300 K gas. At this point, the Na atom and all previously deposited rare gas atoms (with the exception of the original 27) were allowed to relax to equilibrium positions. Following the relaxation, the next atom or group of atoms was brought in. A complete simulation run resulted in a Na atom imbedded in a matrix of 150 rare gas atoms.

Relaxation proceeded in the following way. At the  $n$ th step of the relaxation process, the program calculated new positions  $\mathbf{r}_i(n)$  for the rare gas atoms and a new Na position  $\mathbf{R}(n)$  according to the relations

$$\mathbf{r}_i(n) = \mathbf{r}_i(n-1) - (\nabla_i V) \delta r / |\nabla_i V|, \quad (2)$$

and

$$\mathbf{R}(n) = \mathbf{R}(n-1) - (\nabla_R V) \delta r / |\nabla_R V|, \quad (3)$$

where  $\nabla_i$  and  $\nabla_R$  are gradients with respect to the coordinates of the  $i$ th rare gas atom and the coordinates of the Na atom, respectively. The step size  $\delta r$  was the same for all atoms.

The relaxation iterations proceeded as long as

$$V[\mathbf{r}_1(n), \dots, \mathbf{r}_N(n), \mathbf{R}(n)] \leq V[\mathbf{r}_1(n-2), \dots, \mathbf{r}_N(n-2), \mathbf{R}(n-2)]. \quad (4)$$

We found empirically that seeking the minimum of the

system's potential energy by comparing  $V(n)$  to  $V(n-2)$ , rather than to  $V(n-1)$ , invariably carried the system over pseudominima in the potential.

The initial step size  $\delta r$  was large (0.5 Å), but each time the condition of Eq. (4) was violated, which meant the last iteration had overshoot the potential minimum, the step size was reduced. The final equilibrium positions of the atoms at the end of each relaxation stage in the deposition process were obtained with a step size of 0.03 Å.

This simulation program is less sophisticated than full-fledged molecular-dynamics programs conventionally used to simulate crystal growth, but it consumes far less computer time and, as will be discussed in the next section, it yielded plausible and interesting results.

## B. Calculation of absorption band wavelengths

Once a simulated trapping site had been obtained, the methods of Ref. 8 were used to calculate the matrix-perturbed energy levels of the Na ground state and lowest  $p$  state. The energy difference of the Na states in a particular site gave a theoretical value for the central absorption peak wavelength. The Na-rare gas dimer potentials for the Na  $p$  state are not simply additive, and, as discussed in Ref. 8, the perturbation is obtained by a matrix diagonalization involving the  $A^2\Pi$  and  $B^2\Sigma$  potentials.

## C. Simulation of excited-state relaxation

Because the Na-rare gas interaction changes radically when the Na atom is optically excited to the  $p$  state, the equilibrium positions of the excited Na atom and its rare gas neighbors can be quite different from their positions when the Na atom is in its ground state. Movement of the Na atom and its neighbors in response to the changed potential can result in a substantially red-shifted emission.

To simulate motional relaxation, we used the relaxation program utilized to generate the trapping sites, but in place of the additive  $X^2\Sigma$  potentials, which generate the potential  $U_i(\mathbf{r}_i, \mathbf{R})$  in Eq. (1), we used the  $p$ -state Na-rare gas potential calculated by the method of Ref. 8. Starting with the ground-state equilibrium positions for a particular site, the program allowed the Na atom and all the rare gas atoms to follow the gradients of the new system potential until new equilibrium positions were reached.

## D. Calculation of the emission band wavelengths

Following the simulated relaxation of the Na atom and the rare gas atoms in response to the changed Na-rare gas potential, the perturbed Na ground-state and  $p$ -state energy levels were recalculated for the new equilibrium positions. The energy difference yielded a theoretical value for the peak emission wavelength corresponding to each simulated trapping site.

## III. RESULTS

As a preliminary test of the simulation program we grew Ar and Xe crystals without the presence of a Na impurity atom. Simulated depositions resulted in both fcc and hcp crystals, with occasional single-atom vacancies and/or

stacking faults. This is completely consistent with x-ray analyses<sup>16,17</sup> of actual rare gas crystals. Although fcc is the dominant phase, the energy advantage over the hcp phase is slight, and both phases have been observed. Experimentally, vacancies are numerous, as are stacking faults, particularly in samples where the hcp and fcc phases coexist.

### A. Xe-Na

We were particularly interested in discovering whether or not the computer simulations would trap Na atoms in substitutional sites. Ossicini and Forstmann<sup>7</sup> have suggested, on the basis of their pseudopotential calculations, that the thermally stable blue-shifted  $3s-3p$  absorption band of Na in Xe corresponds to a substitutional site. If substitutional sites are formed at all for Na in rare gas solids, they are most likely to be formed in Xe, as the Xe-Na equilibrium separation (5.0 Å) is only slightly larger than the Xe-Xe internuclear distance (4.36 Å). Furthermore, the blue-shifted triplet absorption band in Xe is markedly sharper<sup>3</sup> than the other bands in Xe or in Kr or Ar, indicating a highly stable site with a well-defined structure.

Indeed we found that substitutional sites were very readily formed in our simulations, for both hcp and fcc structures. Simulations of relaxation in the excited state resulted in the movement of the Na atom to the side of the matrix cage, with little distortion of the surrounding matrix.

Calculations of the Na energy-level structure in the initial and excited-state positions of the Na and Xe atoms yielded an absorption wavelength of 5400 Å and an emission wavelength of 8300 Å. These results were the same for both the hcp and fcc crystal structures. Experimentally,<sup>3,5</sup> the central absorption peak of the Na-Xe blue triplet is located at 5600 Å and the emission peak occurs at 8500 Å. Given the uncertainties in existing dimer potentials, this agreement is most satisfactory.

The symmetry of the substitutional site resulted in degenerate  $p$ -state energy levels, and one must appeal to the Jahn-Teller effect to explain the observed triplet splitting. This explanation of the splitting is consistent with the MCD spectral studies of Na in Xe<sup>5</sup> and of matrix-isolated group II atoms<sup>2</sup> and is carefully discussed in Refs. 2 and 5.

Only one other type of trapping site was consistently formed in Xe in the computer simulations, and it was a site which destroyed the crystal symmetry in the vicinity of the Na atom. The site evolved from the beginnings of a substitutional site and was formed as a result of Xe atoms coming into positions corresponding to a superposition of both fcc and hcp phases. The configuration of this site is shown in Fig. 2. Interestingly, the local symmetry, although no longer that of the crystal, is high enough to result in a degeneracy of the  $p$  levels. This site had a larger volume than the substitutional site. The Na atom was surrounded by 16 nearest neighbors, rather than 12, and the average Na-Xe distance was 4.7 Å.

Although this second site is not a simple vacancy in an otherwise regular crystal, it is hardly amorphous, and one might expect it to exhibit greater thermal stability than the sites responsible for the red triplet absorption in Xe. The calculated absorption and emission wavelengths for this site

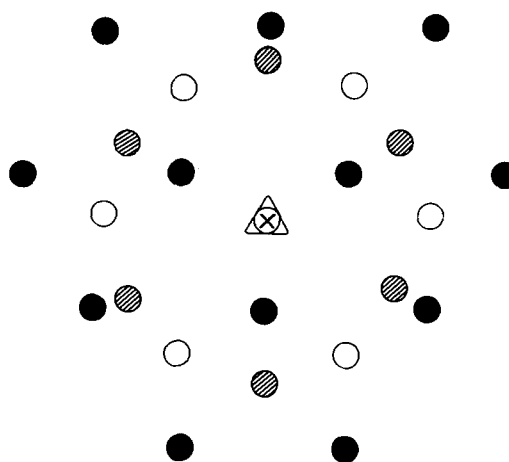


FIG. 2. A top view of the second type of site formed in simulations of the trapping of Na in a Xe matrix. The site evolves out of a substitutional site and is structured in a way that leaves the perturbed Na  $p$  levels degenerate. The black dots indicate the positions of Xe atoms that have deposited to form a (111) plane above the substrate. The Na atom, indicated by a circle with an X, and the open circles, representing the next layer of Xe nearest neighbors are in positions corresponding to a substitutional site. The nearest neighbor Xe atoms in the third layer, however, have come into positions corresponding to both fcc and hcp packing, and the Na-Xe interactions has pulled them inward toward the Na atom at the effective center of the site. These Xe atoms are represented by shaded dots. The last Xe nearest neighbor is indicated by a triangle, and this atom is located directly above the Na atom at a height approximately equal to the height of the fourth layer of Xe atoms in the matrix. The translational symmetry of the lattice is broken by this site geometry.

are 5200 and 7500 Å, respectively. Experimentally,<sup>3</sup> optical excitation of the blue absorption triplet produces a distinct but smaller emission peak at 7100 Å, and there are definite experimental indications<sup>4</sup> that the presence of more than one peak in the emission is due to distinct trapping sites with overlapping absorption bands. Bearing in mind the uncertainties in the Na-Xe dimer potentials, the present results are consistent with the supposition that this second simulated trapping site is responsible for the secondary emission peak associated with optical excitation of the blue triplet.

As a test of the sensitivity of the calculations to the particular Na-rare gas potentials used, we varied both the  $B^2\Sigma$  and  $A^2\Pi$  Na-Xe potentials<sup>12</sup> and calculated the resulting changes in the absorption and emission wavelengths for a substitutional site. Reducing the  $B^2\Sigma$  potential by 15% left the absorption wavelength essentially unchanged but increased the emission wavelength from 8300 to 10 000 Å. The calculations were also sensitive to the equilibrium separation used for the  $A^2\Pi$  state. The Laskowski<sup>12</sup> potential's equilibrium separation is 3.31 Å. Increasing it to 3.6 Å increased the absorption wavelength to 5900 Å and decreased the emission wavelength to 7700 Å. Reducing the equilibrium separation to 3.0 Å produced absorption and emission wavelengths of 5000 and 13 000 Å, respectively. Clearly the results are quite sensitive to the potentials employed in the calculations, but it is also clear that an arbitrary alteration of Laskowski's potentials leads to poorer agreement between experiment and theory.

### B. Na-Ar

The simulations for Na in Ar produced dramatically different results from those obtained for Xe. The Na-Ar

equilibrium separation in 4.99 Å, which is considerably larger than the 3.76 Å Ar–Ar internuclear distance, making it difficult to form substitutional sites. Not one simulation resulted in the formation of a substitutional site. Indeed, in many simulation runs the Na atom was not successfully trapped during deposition; it was squeezed to the surface of the growing matrix as the Ar atoms were deposited. This is consistent with the experimental observation that Na absorption bands grow much more quickly in Xe than in Ar.

The apparent improbability of substitutional site formation has been discussed in previous papers<sup>6,7</sup> reporting calculations of Na spectra in Ar, and larger trapping sites consisting of two-, three-, and four-Ar vacancies in a regular fcc lattice have been considered. The principal theoretical difficulty with two- or three-Ar vacancies in an otherwise regular lattice is that these sites have a pronounced axial symmetry which automatically results in a large doublet splitting<sup>6,8</sup> of the Na *p*-state energy levels on the order of 5000 cm<sup>-1</sup>, and as yet there is no clear experimental evidence of such a widely split doublet absorption. The four-Ar vacancy, on the other hand, has a symmetry which produces degenerate *p* levels, and such a site was considered in Refs. 6 and 7 as a possible candidate for the Na–Ar blue-shifted triplet absorption.

The simulations, however, resulted in essentially amorphous trapping sites, no two of them identical. The coordination numbers of the sites ranged from 15 to 18, but all of the sites had one characteristic in common: they evolved from a two-Ar vacancy in the (111) plane in which the Na atom was imbedded, and the effect of the Na–Ar interaction was to distort this vacancy and force Ar atoms coming in on top into an arrangement with an overall local symmetry sufficient to produce degeneracy in the matrix-perturbed Na *p* levels.

Despite the apparent differences in these sites, calculations of the corresponding absorption and emission wavelengths yielded essentially identical results: an absorption wavelength of 5700 Å and an emission wavelength of 7100 Å. Because of the amorphous nature of these simulated sites, we compare these calculated wavelengths with the spectra associated with the thermally unstable red triplet for Na in Ar. Experimentally,<sup>3</sup> the absorption triplet is centered at 5875 Å and the emission peak occurs at 7000 Å. The equivalent experimental wavelengths for the thermally stable blue triplet are 5450 and 6800 Å. Thus, the calculated wavelengths do appear to correspond most closely to the spectra of the red triplet.

Our simulation program produced no obvious candidates for the site or sites responsible for the stable blue triplet. Yet such a site exists. Both a substitutional site and a four-Ar vacancy possess the symmetry required to produce degenerate *p* levels and both types of sites might be expected to be thermally stable. We therefore decided to force the Na atom into a substitutional site in an initially regular crystal, and into a four-atom vacancy, and to calculate the corresponding absorption and emission wavelengths. In a previous paper<sup>8</sup> we used Na–Ar potentials to calculate the perturbed energy-level structure of Na in one-, two-, and four-Ar vacancies in regular crystals, but the Na atom was

placed within undistorted vacancies and the red shift of the emission was calculated by permitting only the Na atom to move in response to the change in the Na–Ar potential following optical excitation. With our relaxation program in hand, it was possible to recalculate the spectra in a more realistic fashion.

We placed the Na atom in a substitutional site and in a four-Ar vacancy in a regular fcc crystal consisting of 100 Ar atoms and then allowed the whole system to relax with the Na atom in the ground state. For the substitutional site, this resulted in a slightly expanded trapping-site volume. We calculated an absorption wavelength and then allowed the entire system to relax with the *p*-state Na–Ar potential in order to calculate the emission red shift. Our calculated absorption and emission wavelengths were 5700 and 6750 Å for the substitutional site and 5700 and 6470 Å for the four-Ar vacancy. The experimental wavelengths for the blue triplet are 5450 and 6800 Å. Thus, although it is not obvious from our growth simulations that a substitutional site can be formed, the wavelength calculations favor an identification of the blue triplet with a substitutional site rather than with a four-Ar vacancy.

#### IV. CONCLUSIONS

The present results strongly support the assignment of the blue absorption triplet for Na in Xe to a substitutional site. Not only were such sites regularly formed by computer simulations, but the wavelengths for the absorption and emission bands calculated on the basis of existing Na–Xe dimer potentials are in excellent agreement with experiment. In addition, a second structured site formed in the simulations is a candidate for the site responsible for the weaker short wavelength emission peak associated with optical excitation of the blue triplet.

The computer simulations of trapping site formation for Na in Ar, on the other hand, offer a ready explanation of the thermally unstable Na–Ar red triplet. In this picture, the red triplet is the result of overlapping absorption and emission bands due to numerous amorphous sites with varying local configurations and coordination numbers, all of which produce approximately the same perturbation of the Na energy levels. The calculated absorption and emission wavelengths corresponding to these simulated sites do overlap, and they agree quite well with the experimental data for the red triplet.

The simulations for Ar did not produce sites which could be associated with the stable blue triplet, but forcing the Na atom into a substitutional site and allowing the system to relax resulted in a trapping site with calculated wavelengths approximating those for the blue triplet. Similar calculations for a regular four-Ar vacancy produced less satisfactory results.

One is drawn to the inference that the simulation program erred on the side of regular, highly structured trapping sites in the case of Xe and on the side of amorphous sites for Ar, and that reality lies somewhere in between. This view leads one to a particularly simple explanation of the origin of the principal absorption and emission bands of matrix-iso-

lated Na: the blue triplet is due to substitutional sites, with possible contributions to the emission from closely related, structured sites deviating from the substitutional geometry due to a local mixing of fcc and hcp phases; and the red triplet is due to the superposition of bands corresponding to amorphous trapping sites which evolve from two-atom vacancies.

A highly interesting feature of the simulations was the fact that even in the case of amorphous sites, the local symmetry was high enough to produce a degeneracy of the perturbed Na  $p$  levels. This favors the Jahn–Teller effect as the explanation of the observed splitting of the absorption bands. The nonsubstitutional sites formed in the computer simulations, however, lacked the translational symmetry exhibited by a crystal. Only the substitutional site could be characterized by a standard crystal symmetry.

<sup>1</sup>H. Kuppelmaier, H. J. Stoeckmann, A. Steinmetz, E. Goerlach, and H. Ackerman, *Phys. Lett. A* **98**, 187 (1983).

- <sup>2</sup>J. Hormes and J. Schiller, *Chem. Phys.* **74**, 433 (1983).
- <sup>3</sup>L. C. Balling, M. D. Havey, and J. F. Dawson, *J. Chem. Phys.* **69**, 1670 (1978).
- <sup>4</sup>J. J. Wright and L. C. Balling, *J. Chem. Phys.* **73**, 994 (1980).
- <sup>5</sup>J. C. Miller, R. L. Mowery, E. R. Krausz, H. W. Kim, P. N. Schatz, and L. Andrews, *J. Chem. Phys.* **74**, 6349 (1981).
- <sup>6</sup>J. F. Dawson and L. C. Balling, *J. Chem. Phys.* **71**, 836 (1979).
- <sup>7</sup>S. Ossicini and F. Forstmann, *J. Chem. Phys.* **75**, 2076 (1981).
- <sup>8</sup>L. C. Balling and J. J. Wright, *J. Chem. Phys.* **79**, 2941 (1983).
- <sup>9</sup>J. Tellinghuisen, A. Ragone, M. L. Kim, D. J. Auerbach, R. E. Smalley, L. Wharton, and D. H. Levy, *J. Chem. Phys.* **71**, 1283 (1979).
- <sup>10</sup>G. York, R. Scheps, and A. Gallagher, *J. Chem. Phys.* **63**, 1052 (1975).
- <sup>11</sup>R. P. Saxon, R. E. Olson, and B. Liu, *J. Chem. Phys.* **67**, 2692 (1977).
- <sup>12</sup>B. C. Laskowski, S. R. Langhoff, and J. R. Stallcop, *J. Chem. Phys.* **75**, 815 (1981).
- <sup>13</sup>W. E. Baylis, *J. Chem. Phys.* **51**, 2665 (1964).
- <sup>14</sup>J. A. Barker, R. A. Fisher, and R. O. Watts, *Mol. Phys.* **21**, 657 (1971).
- <sup>15</sup>J. A. Barker, R. O. Watts, J. K. Lee, T. P. Schafer, and Y. T. Lee, *J. Chem. Phys.* **61**, 3081 (1974).
- <sup>16</sup>C. S. Barrett and L. Meyer, *J. Chem. Phys.* **41**, 1078 (1964).
- <sup>17</sup>S. I. Kovalenko, E. I. Indan, A. A. Solodovnik, and I. N. Krupskii, *Sov. J. Low Temp. Phys.* **1**, 493 (1975).